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SYSTEMS ANALYSIS REQUIREMENTS FOR NITROGEN OXIDE CONTROL OF STATIONARY SOURCES

Aerotherm/Acurex Corporation

Prepared for
National Environmental Research Center

September 1974

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The report gives results of a study to identify systems requirements for the control of nitrogen oxide (NOx) emissions from stationary sources. It evaluates developments in the character of NOx emission sources and in NOx control technology. It is concluded that planning priority should be for coal-fired utility and industrial boilers, followed by stationary internal combustion (I.C.) engines. The most attractive short- and long-term option for control of NOx emissions is combustion modification technology. The priority items are development of techniques for control of the conversion of fuel-bound nitrogen to NO, and development of combustion modifications for the major area sources such as pipeline I.C. engines, and commercial and domestic combustion units.

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FOREWORD

This document presents the results of a brief systems study of nitrogen oxide control methods for stationary sources. The report provides updated information on NO_x emission sources and NO_x control technology and concludes with recommendations of R&D requirements.

Aerotherm extends its appreciation for the valuable assistance provided by the following individuals, organizations, and companies: Dr. J.O.L. Wendt and D.L. Pershing of the University of Arizona; C.R. McCann of the U.S. Bureau of Mines; W.H. Barr of the Pacific Gas and Electric Co.; R.N. Levine and K.A. Krumwiede of the Southern California Edison Co.; W.J. Armento and W.L. Sage of the Babcock and Wilcox Co.; D.P. Teixeira of the Electric Power Research Institute; W. Bartok and A. R. Crawford of Esso Research and Engineering. We would also like to give special thanks for information provided by the following members of the Control Systems Lab of EPA: J. S. Bowen, G. B. Martin, R. E. Hall, D. B. Lachapelle, W. S. Lanier, J. H. Wasser of the Combustion Research Section; Bruce Henschel of the Advanced Process Section and G. Haselberger of the Regenerable Processes Section.

This survey was performed for the Engineering Analysis Branch of the Control Systems Laboratory, U.S. Environmental Protection Agency. R.F. Sanders was the task officer. The Aerotherm Project Manager was Dr. Larry W. Anderson. Dr. Carl B. Moyer acted as advisor for all phases of the study. The study was performed during the months of January and February, 1974.

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SECTION 1

SUMMARY

This report presents the results of a two-month study by the Aerotherm Division of Acurex Corporation to identify systems requirements for the control of nitrogen oxide emissions from stationary sources. The study focused on the evaluation of developments, since the 1969 Esso Research and Engineering Systems study[1], in the character of NO_x emission sources and in NO_x control technology. It is concluded that planning priority should be for coal fired utility and industrial boilers followed by stationary internal combustion engines. The most attractive short- and long-term option for control of NO_x emissions is combustion modification technology. The priority items are development of techniques for control of the conversion of fuel-bound nitrogen to NO, and development of combustion modifications for the major area sources such as pipeline I.C. engines, and commercial and domestic combustion units.

1.1 CHARACTERIZATION OF EMISSION SOURCES

Currently available inventories of NO_x emission were compared with the data presented in the 1969 Esso study. No new major sources of NO_x were discovered. Table 1-1 shows a comparison of the Esso projected inventory for 1970 with two recent EPA inventories (References 2 and 3). Figure 1-1 presents the Reference 2, AP-115, data as a pie chart. As can be seen from the data the greatest difference is in the utility-size boilers category which increased to 4.7×10^6 tons/year from the earlier estimate of 3.84×10^6 tons/year. This difference is due to more refined analysis of boiler types and their individual emission factors as well as an updated emission factor for gas-fired boilers. Reference 3 indicates a further increase in utility boiler emissions; however, these data are rather preliminary.

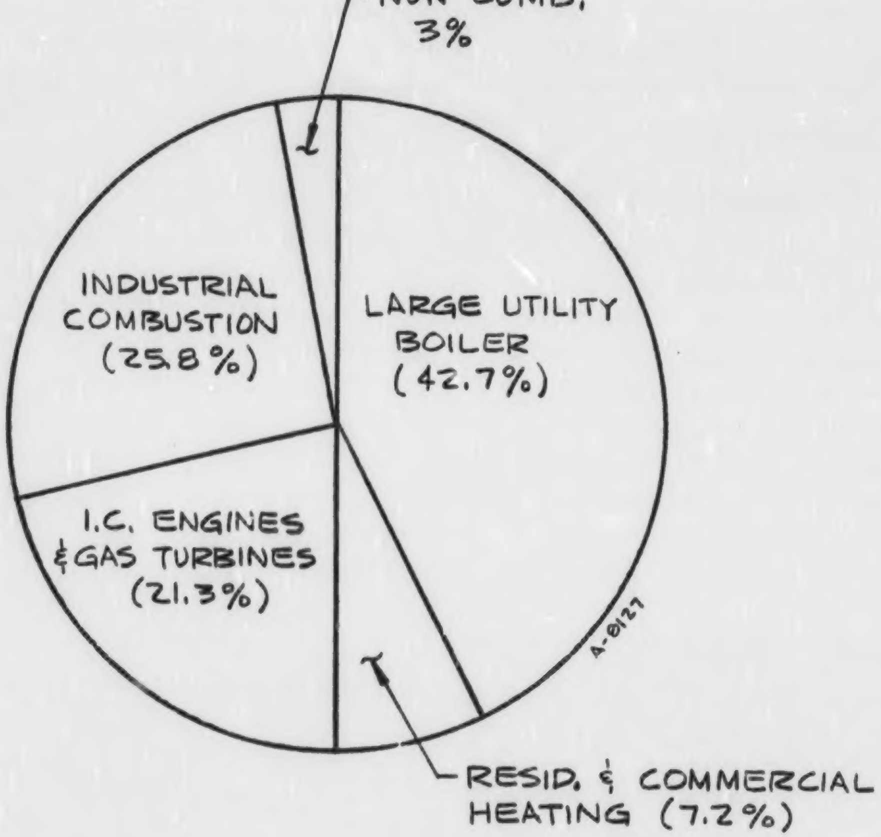
A further breakdown of the emission sources reveals that, since the Esso study, coal combustion has remained as the largest contributor to the total stationary source NO_x emissions at 42 percent. In electrical utility generation, coal contributes 63 percent of the NO_x emissions. It is anticipated that the fractional contribution and total levels from coal will increase substantially during the next decade.

TABLE 1-1

TOTAL NO_x EMISSIONS FROM STATIONARY SOURCES (X 10^6 tons/year)

Source	EPA (1974)(3)	AP-115, 1971(2)	Esso, est. 1970,(1)	Other, 1970(3,4,5)
Utility Size Boilers	5.88	4.71	3.841	3.3-6.0[c]
Recip. I. C. Engines	0.918	4.53	2.097[b]	1.95-2.3
Industrial Size Boilers	3.91		2.81	
Forest Fires (controlled)	d	.2 75	a	
Domestic And Commercial Space Heaters	0.945	0.79	1.001	
Gas Turbines	d	a	a	
Solid Waste Disposal	0.179	0.395	a	
Process Heaters	2.876	a	a	
Nitric Acid Plants	d	a	0.235	
Other Sources	d	0.338		
Total	14.7	11.038	9.986	

Notes:
- a. Included In Industrial-size Boilers
- b. Pipeline And Gas Plants Only
- c. Depending on Load Factor (High Number For Full Load)
- d. Not included in NEDS data



NO_x ESTIMATED TONS, 1971	SOURCE CATEGORY
4,710,000	LARGE UTILITY BOILERS
2,350,000	I.C. ENGINES & GAS TURBINES
2,850,000	INDUSTRIAL COMBUSTION
790,000	RESIDENTIAL & COMMERCIAL HEATING
340,000	NON-COMBUSTION SOURCES
11,040,000	

Figure 1-1. Total NO_x Emitted in the U.S. from Stationary Sources, 1971 (From Data Obtained from References 2 and 4)

Figure 1-1 reveals that stationary I.C. engines and gas turbines contribute 21 percent to the total NO_x emissions. There is a considerable range of estimates of NO_x emissions from I.C. engines starting from a low of 0.95 x 10^6 tons/year to a high of 2.3 x 10^6 tons. The effect of these emissions on overall air quality is unknown.

1.1.1 Emission Factors

Emission factors have been updated and more explicitly defined for each source category. A few sources which appear in the Esso report are absent from the latest EPA emission factor data book, AP-42. These include diesel I.C. engines installed in petroleum refineries, boilers and natural gas engines in natural gas plants, natural gas engines in pipelines, sintering and other furnaces in steel-making, lime kilns and glass manufacture. Many of these undelineated topics may be imbedded in other categories. Recent data show emission factors for oil fired residential heaters 67 percent higher than reported by Esso. These latest emission factors for residential units have not been incorporated in the reported emission inventories shown in Table 1-1.

1.1.2 Prospects for NO_x Emission

Trends for the future include the impact of the following factors:

- Changes in energy demand
- Fuel switching
- Adaptation of controls.

Figure 1-2 shows a long term projection of fuel utilization for electrical utility generation[8]. This curve assumes that fuels will be readily available and that generating plants will be built to meet the demand. Environmental laws and international political developments have hindered these sources in the recent past. Thus we see a much greater utilization of direct coal combustion in the next decade. From 1980 to 2000 there will be greater emphasis on "clean fuels" derived from coal, i.e., synthetic gas and oil.

For residential and commercial sources we see the following trends:

- Reduced emissions due to reduced load (cooler homes)
- Some switching back to coal-fired home heating systems
- In the longer term, switching to electrical heat (provided the power plants can meet the demand)
- Increases in efficiency of the home heater system and greater housing insulation to reduce the total load and thus emissions

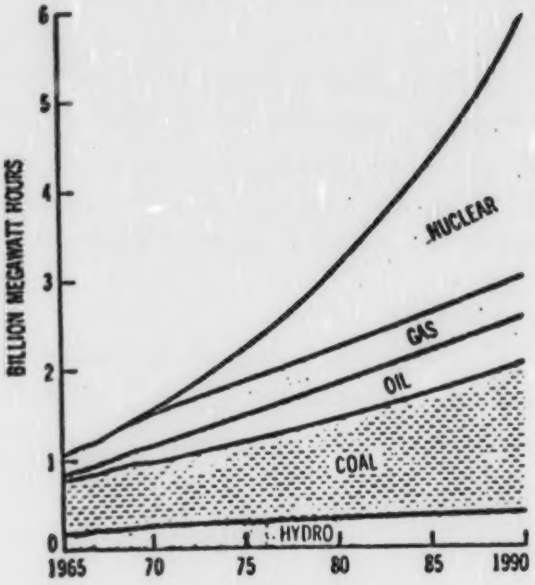


Figure 1-2. Estimated Annual Electric Utility Generation by Primary Energy Sources

- By the year 2000, the possible widespread use of solar heating for homes.

Total estimated NO_x emissions from industrial combustion has not changed much since 1969. Little field testing has been done in this area and, except for large industrial boilers, control techniques have not been well defined. Emission control efforts will continue to expand, along with fuel substitution trends.

Control schemes for the primary non-combustion source, nitric acid plants, have been developed to meet 1980 EPA standards.

Total stationary source NO_x trends have exceeded the predicted trends as shown in Figure 1-3. However, there is a slight downward trend due to revised emission estimates and controls on West Coast utilities.

1.2 SURVEY OF OPTIONS FOR NO_x CONTROL

Since the Esso study the effectiveness of numerous NO_x control options has been evaluated. Present or potential options are categorized as follows:

- Modification of the existing process
- Modification of the fuel
- Treatment of the flue gas
- Use of an alternate process.

These options were evaluated in this report on the basis of the following criteria:

- Cost effectiveness
- Availability (short-term options)
- Low risk development (long-term options)
- Impact on unit efficiency
- Impact on operational difficulty
- Overall environmental impact due to process control
- Compatibility with projected fuel allocations.

The results are summarized in Table 1-2. For the control of existing combustion sources, combustion modification has remained the most expedient, most reliable and cost effective technique. For noncombustion sources, flue gas treatment is preferred.

For long term NO_x control, the incorporation of combustion modifications into new unit design appears to be the single most effective strategy for

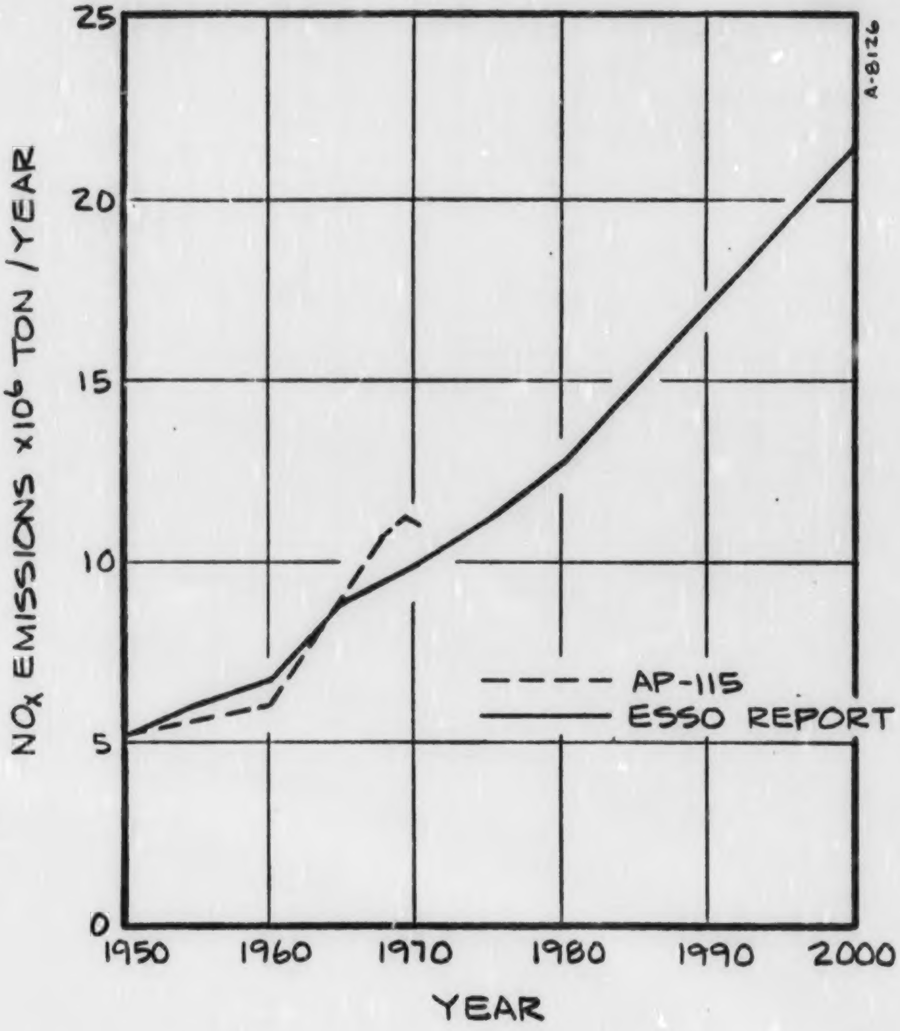


Figure 1-3. Stationary Source NO$_x$ Emission Trends

TABLE 1-2

SUMMARY OF NO$_x$ CONTROL OPTIONS

Option	Approach to NO$_x$ Control	Effectiveness		Overall Evaluation
		Short Term	Long Term	
Combustion Modification	Suppress formation of thermal, fuel No$_x$ by modifying operating conditions	Present applicability; preferred option	Incorporate into optimized low NO$_x$ design	Priority option for short and long term control of boilers, furnaces, turbines
Synthetic Fuels	Reduced flame temperatures from low Btu gas yields reduced NO$_x$	Negligible application	Promising low risk concept for post-1980	Attractive long term option; May require combustion modifications
Fuel Additives	Reduce or decompose NO to N$_2$ by catalytic addition	Not effective	High risk development	Not promising
Fluidized Bed Boiler	Low bed temperature suppresses thermal NO$_x$	N.A.	Medium risk concept for post-1980	Promising long term option; May require combustion modifications
Flue or Exhaust Gas Treatment	Reduce NO to N$_2$ by catalytic treatment	Preferred for nitric acid plants	Possible for control of I.C. Engines	Potential long-term option for backup of combustion modification for boilers, furnaces. Possible long-term option for I.C. Engines. Priority option for nitric acid plants.

stationary sources. Large electric power generation units will undoubtedly benefit from developments in the use of synthetic fuels and fluidized bed combustion in conjunction with combined power cycles. Even these advanced concepts, however, may require combustion modification for NO_x control. The bulk of stationary sources -- area sources such as pipeline I.C. engines, industrial combustion, and domestic heaters -- will increasingly utilize combustion modifications for NO_x control in new equipment design.

We conclude that combustion modifications are clearly the priority item in program planning for NO_x control of stationary sources.

1.3 COMBUSTION MODIFICATION TECHNOLOGY

Since the Esso study, there have been fundamental advances in identifying the formative mechanisms for thermal and fuel NO_x. The capability for relating NO_x formation mechanisms to specific combustion modifications has not yet been established, however. Accordingly, the EPA program to devise and test modification techniques is broadly based with simultaneous focus on fundamental studies, pilot scale testing on laboratory scale equipment, and full scale testing on commercial equipment. The overall short term objective of the program is to develop verified, cost effective techniques for retrofit modification of existing units. Most of the significant reductions in NO_x emissions have resulted or directly benefited from the efforts of this program. The overall long term objective of the program is to develop optimized control concepts for new units and to define minimum attainable emission levels. For both the short and long term studies, the priority development area is combustion modification techniques for control of fuel nitrogen conversion to NO.

1.3.1 Fundamental Studies

The fundamental studies area of the combustion control program is primarily a long term effort to develop a basic understanding of NO_x formation. Since the Esso study, the emphasis has been on modeling the individual key phenomena involved in NO_x formation. Significant advances have been made in the areas of fluid flow solution, thermal NO_x kinetics, gaseous hydrocarbon oxidation, and thermal radiation. These results have had the short term yield of aiding in evaluating data from pilot and full scale tests, and in suggesting ideas for combustion modification techniques. Only preliminary advances have been made in the modeling of fuel NO_x and of the effects of turbulent mixing on hydrocarbon and NO reaction rates. These are both considered priority R&D areas. In the long term, the individual models for the key phenomena will be

TABLE 1-3a

MODIFICATION TECHNIQUES FOR UTILITY AND INDUSTRIAL BOILERS

Modification Techniques	Major Effect	Major Potential Problems
Utility and Industrial Boilers		
Low Excess Air Operation	Oxygen concentration reduction	Increased furnace slagging Nearer smoke threshold
Off-Stoichiometric Combustion	Fuel-rich burner operation Reduced residence time of fuel at peak temperature Peak temperature reduction	Flame instability, smoking Higher convective section temperatures
Flue Gas Recirculation	Peak temperature reduction	Flame instability Boiler efficiency reduction
Reduced Air Preheat Operation	Peak temperature reduction	Boiler efficiency reduction
Load Reduction	Peak temperature reduction	Boiler efficiency reduction
Water, Steam Injection	Peak temperature reduction	Boiler efficiency reduction Increased corrosion
Equipment Modifications:		
● "Low NO_x" Burner	Provides off-stoichiometric combustion at burner Readily fuel/air adjustable Fuel-flexible	Retrofit problems: Compatibility with existing furnace Conversion investment costs Boiler downtime
● Boiler Wash	Maintains rated heat transfer rate	Boiler downtime Maintenance expense
● Widen Burner Spacing	Reduce peak temperature Reduce interference between burner flames	New unit design only; retrofit cost prohibitive
● Tangential firing (as opposed to wall-firing)	Peak temperature reduction Slower, more controlled, combustion	

TABLE 1-3b

MODIFICATION OR CONTROL TECHNIQUES FOR STATIONARY I.C. ENGINES AND GAS TURBINES

Modification or Control Techniques	Major Effect	Major Potential Problems
Reciprocating I.C. Engines		
Speed vs. Stoichiometry	With speed, NO increases under fuel-rich and decreases under fuel-lean conditions	Retrofit difficulties; inability to meet load demand
Decreased Torque Load (at constant speed)	Peak temperature reduction	Retrofit difficulties; inability to meet load demand
Decreased Air Manifold Temperature	Peak temperature reduction	Efficiency reduction
Increased Valve Overlap	Peak temperature reduction	Fuel economy reduction Applicable only to 4-cycle engines
Exhaust Gas Recirculation	Peak temperature reduction	Intake manifold fouling Additional control system Operational difficulties Efficiency reduction
Catalytic Converter, Ammonia as Reducing Agent (Post Combustion Control Method)	Reduction of NO, NO_2 to N_2 and O_2	Expensive Current catalysts sensitive to impurities in fuel
Water Injection	Peak temperature reduction	Increased maintenance; additional equipment for water handling
Precombustion chamber	Peak temperature reduction; O_2 starvation	Costly for retrofit

TABLE 1-3b (Concluded)

Modification or Control Techniques	Major Effect	Major Potential Problems
Gas Turbines		
Lean-Out Primary Zone by Modifying Combustion Chamber Design	Peak temperature reduction Reduced residence time of fuel at peak temperature	Less control over flame stabilization Less control over lower lean extinction performance
Water injection	Peak temperature reduction	Reduced efficiency Increased maintenance Additional equipment for water handling
Exhaust Gas Recirculation	Peak temperature reduction	Reduced efficiency Additional control system Operational difficulties

coupled, refined, and verified by correlation with test data to ultimately yield predictions of NO_x formation.

1.3.2 Application of Combustion Modifications

The present study concludes that combustion modification, including both operation and equipment variation, is the most viable means of reducing NO_x formation from stationary sources. Tables 1-3a and 1-3b list the most common modification techniques applicable to utility and industrial boilers, stationary gas turbines and reciprocating I.C. engines.

The effectiveness of these combustion modifications has been demonstrated more completely during the past several years. The record of achievement on steam electric power plants is particularly impressive, with response to these modifications dependent mainly on fuel type. NO_x from gas-fired installations showed the greatest reductions since thermal NO_x is the only formation mechanism involved. Diminished degrees of success have been attained for oil and coal firing due to the influence of fuel NO_x.

For all three fuels, off-stoichiometric combustion techniques have proven to be the most effective in suppressing both thermal and fuel NO_x. Firing under low (5-10 percent) excess air conditions has become standard operating procedure for most utility and industrial boilers. The other modification methods, such as flue gas recirculation and reduced air preheat, are increasingly costly to implement in terms of investment, operating costs, and loss of plant efficiency.

Reports have shown that serious operating problems theoretically associated with combustion modifications can largely be avoided. However, a certain amount of additional vigilance, as well as willingness to tolerate a decrease in flexibility, is required of the plant operator.

Since 1969, a lesser amount of practial NO_x modification work has been performed on stationary reciprocating I.C. engines and gas turbines. This is due in part to a general lack of appreciation of the importance of these sources. The contribution of this type of source has since been estimated to be quite significant. The most promising NO_x reduction methods include hardware modification for I.C. engines (i.e., precombustion chamber) and water injection for gas turbines.

The combustion modification cost picture is blurred. In most cases, such information has gone unreported or is of a proprietary nature. It has been concluded, however, that generalized cost data would be difficult to compile due to the disparity between the responses of essentially identical combustion systems to a given modification. These disparities are due to plant condition, operating modes, fuel differences, etc.

The current clean fuels shortage has significantly influenced the course of NO_x control programs for utility and industrial boilers. The encouraging results obtained from natural gas and oil firing have become somewhat academic due to the probable widespread substitution of coal for these fuels. It is therefore mandatory that all aspects of combustion modification techniques for coal firing be thoroughly researched in both sub- and full-scale tests. Achieving maximum NO_x suppression and thermal efficiency concurrently are the ultimate goals.

As emissions from utility and industrial boilers are controlled, the contributions from area sources, such as stationary reciprocating I.C. engines and gas turbines, will become more significant. NO_x control systems R&D and their application to on-line systems is currently accelerating.

1.4 ASSESSMENT OF R&D REQUIREMENTS

Table 1-4 summarizes the items identified in this study as requiring further development. The recommendations are categorized as follows:

- Combustion Modification Technology
    - Fundamental Studies
    - Fuels R&D
    - Process R&D
    - Field Testing and Survey
- Flue Gas Treatment
- Alternate Processes

The overall priority item is development of control techniques for coal-fired units. Individual items regarded as high priority are summarized below.

- Fuel NO_x - The continuing Rocketdyne study is leading to an understanding of mechanisms by which fuel bound nitrogen is converted to NO. As preliminary results are obtained, independent studies, at a similar level of funding, should be initiated for key aspects of the study such as volatilization and pyrolysis models, and carbon burnout models.
- Mixing Effects on Reaction Rates - An understanding of NO_x formation in practical combustors must account for the effects of distortion of the flame zone due to turbulent mixing. A new program with

TABLE 1-4

RECOMMENDED R&D

R&D Area	Subheading	Program	Priority	New Program/ Continuation	Reasoning
Fundamental Studies	Fluid Flow Solutions	Continue 2-D code development	Medium	Continuation	Incorporate current auxiliary models
		Apply 2-D jet flame boundary layer codes	Medium	New	Test turbulence model, mixing effects on kinetics, 2ϕ flow models, luminous radiation effects
		Pressure-velocity codes for 2-D recirculating flow	Low	New	Utilize current technology
		Pressure velocity method for 3-D	Low	New	Obtain flow patterns and scaling laws
	Thermal NO_x	HCN emission in fuel-rich flames	Medium	Continuation	Determine potential as a pollutant
		Esso-Ultrasystem codes with diffusional effects	Medium	Continuation	Extend findings to practical flow situations
	Fuel NO_x	Volatilization + pyrolysis models	High	Continuation	Extend Rocketdyne findings into independent studies
		Post-pyrolysis heterogeneous carbon burnout models	High	Continuation	" " "
		Couple particulate combustion with luminous radiation	High	Continuation	" " "
		Synergism of NO_x and SO_2	High	Continuation	" " "
		HCN	High	Continuation	Determine potential as a pollutant
	Turbulent Viscosity	2 Equation turbulence models in recirculating 2-D flow code	Medium	New	Utilize current technology
		Nonisotropic turbulence models for swirling 2-D boundary layer jet flames	Medium	New	Verify models in simple flow geometries
	Mixing Effect On Reaction Rates	Experimental and analytical study of turbulent premixed and diffusion flames - simple geometry	High	New	Develop models for effect of mixing on reaction rates
	Thermal Radiation	Coordinate existing radiation models with fuel nitrogen study	Medium	Continuation	Utilize findings of Rocketdyne study
Fuels R&D	Off-Stoichiometric Combustion	Determine optimum design parameters for staged firing with coal	High	New	Present research indicates this to be most viable NOx control technique for coal combustion
	Burner Optimization for Fuel NO_x	Determine optimum design parameters for low NO_x burner	High	New	Effective option for fuel nitrogen control

TABLE 1-4. Continued

R&D Area	Subheading	Program	Priority	New Program/ Continuation	Reasoning
Fuels R&D continued	Burner Optimization for Thermal NO_x	Determine optimum design parameters for low NO_x burner using low nitrogen fuels	High	New	May be effective option for thermal NO_x as well
	Radiation Measurement	Determine realistic radiation heat fluxes for coal, oil and gas	Low	New	Input to radiation models
	Corrosion & Fouling	Determine ability to do subscale corrosion and fouling studies of NO_x control schemes	High	New	Very important factor to determine applicability of controls
	Coal Type	Systematically test different types of coal - effect of properties on NO_x emissions	Medium	New	Determine applicability of control techniques over spectrum of coal types
	Synthetic Fuels	NO_x emissions from fuels and equipment conversion problems	Medium	Continuation	Necessary to identify problems of future fuels
	Oil Atomization	Determine effects of fuel atomization technique on NO_x	Medium	Continuation	Potential control technique; coordinate with optimum burner design
	Optimization Modification	Determine optimized modification schemes for each fuel and process	Medium	Continuation	Necessary for immediate retrofit
Process R&D	Corrosion Studies	Perform full scale studies of NO_x control techniques on corrosion and fouling	High	Continuation	Full scale testing will yield the ultimate data
	Off-Stoichiometric Combustion	Expand full scale testing for staging on coal combustion	High	Continuation	" " "
	Intermediate Bound Compounds	Determine fate of chemically bound nitrogen in full scale equipment	Low	New	Determine if there really is a problem in full scale equipment
	Optimized Modification and Areas Sources	Continue to expand ongoing programs to determine optimized schemes; especially area sources	High	Continuation	Determine applicability on full scale equipment
	Catalytic Combination	Exploratory work to determine full potential	Low	Continuation	Potential long range option
Field Testing	Pulverized Coal-Fired Equipment	Determine current stationary source control technology (CSST)	High	Continuation	Coal-fired equipment has highest priority
	Corrosion & Slagging	Determine effects of CSST on corrosion and fouling	High	Continuation	Additional data very important
	Area Sources	Determine CSST for area sources	High	Continuation	Field data lacking

TABLE 1-4. Concluded

R&D Area	Subheading	Program	Priority	New Program/ Continuation	Reasoning
Field Testing continued	Alternate Fuels	Exploratory work to determine nature and extent of problem	Low	New	Extent of use and NO_x problems are unknown
Flue Gas Treatment	--	Review state of the art	Low	New	May be required for secondary cleanup
Alternate Processes	Synthetic Fuels	Determine NO_x problems	Low	New	Fuels may be extensively used in 1980-2000
	Advanced Power Generation Cycles	Determine NO_x problems	Low	New	" " "

coordinated experimentation and analysis for a simple flow geometry would contribute to the basic understanding of this effect and supplement the coupled modeling effort being done by UARL.

- Off-Stoichiometric Combustion - This is regarded as the most effective retrofit method for simultaneous suppression of thermal and fuel NO_x formation. A new program in fuels R&D and a continuing program in process R&D is required to fully exploit the potential of this technique.
- Burner Optimization - Burner configuration and injection pattern should be optimized, particularly for new unit design, to suppress the formation of thermal and fuel NO_x. A generalized Fuels R&D program to investigate types of firing for gas, oil and coal would provide the data needed for design optimization.
- Corrosion and Fouling - The effectiveness of combustion modification schemes can be limited by the increased propensity for corrosion resulting from the modified combustion conditions. This problem requires study on both subscale and full-scale units.
- Area Source Testing - A field testing program is needed to extend the technology developed for control of utility boilers to the control of the major area sources.

SECTION 2

INTRODUCTION

In 1969, Esso Research and Engineering published results of a system study of NO_x control from stationary sources with the following objectives:

- Characterize stationary source NO_x emissions
- Assess NO_x control technology
- Explore utility of modeling for NO_x predictions
- Analyze cost effectiveness of control techniques
- Recommend R&D plan for NO_x control technology

Esso estimated that stationary sources comprised 60 percent of total NO_x emissions and of that percentage, 98 percent was due to combustion sources ranked as follows: (1) electric power plants (37.5 percent), (2) industrial combustion (29.2 percent), (3) pipelines and gas plants (20.8 percent), (4) domestic and commercial heaters, (10.4 percent). Based on the limited data and experience at that time, Esso concluded that the most promising short term option for NO_x control was combustion modification.

The Esso study was done when the concerted government program for NO_x control was in the preliminary stages. Since that time, there have been numerous developments both in the character of present or projected emission sources and in techniques for NO_x control. Recently, obstacles encountered in achieving effective control of mobile sources have led to a stronger emphasis on control of stationary sources. The supply shortage of petroleum fuels is leading; in the short term, to conversion to coal firing, and, in the long term, to a re-evaluation of alternate combustion processes for replacement of existing units. The effectiveness of the combustion modification strategy for control of large boilers has been validated, since the Esso study, for gas and oil firing, and preliminarily for coal firing. With coal, the strategy is complicated by the conversion to NO_x of the nitrogen compounds in the fuel. Control of the major area sources - industrial, commercial, and domestic combustion units - is being initiated, using where possible, experience derived from control of utility boilers.

In view of the changing pattern of NO_x control for stationary sources, Aerotherm Division of Acurex Corporation has been conducting a study for the Control Systems Laboratory of the Environmental Protection Agency in order to assess the current status of NO_x control technology. Particular emphasis is on identifying changes, since 1969, which would reorient the focus of systems planning. Individual objectives are:

- Characterize current and projected stationary NO_x emission sources
- Evaluate present and potential options for NO_x control
- Characterize the status and prospects of combustion modification technology
- Determine requirements for R&D

The Esso report gave a comprehensive treatment of virtually all aspects of the stationary NO_x emission problem including detailed descriptions of emmission sources and control techniques. For the present study, it was therefore possible to focus, at the outset, on the most significant changes impacting systems planning. Figure 2-1 summarizes the approach used for this study, and, also, provides a schematic outline of this report. The conclusions and recommendations of this study were based on a review of recent literature and discussions with industrial users and manufacturers. Based on the survey of emission sources, given in Section 3, and on the evaluation of control options, given in Section 4, it is concluded that the focus of short-term systems planning remains on combustion modifications for large boilers and the major areas sources. Section 5 is the main section of the report and is devoted to giving an overview of combustion modification technology. Potential R&D areas identified in Section 5 are summarized in Section 6.

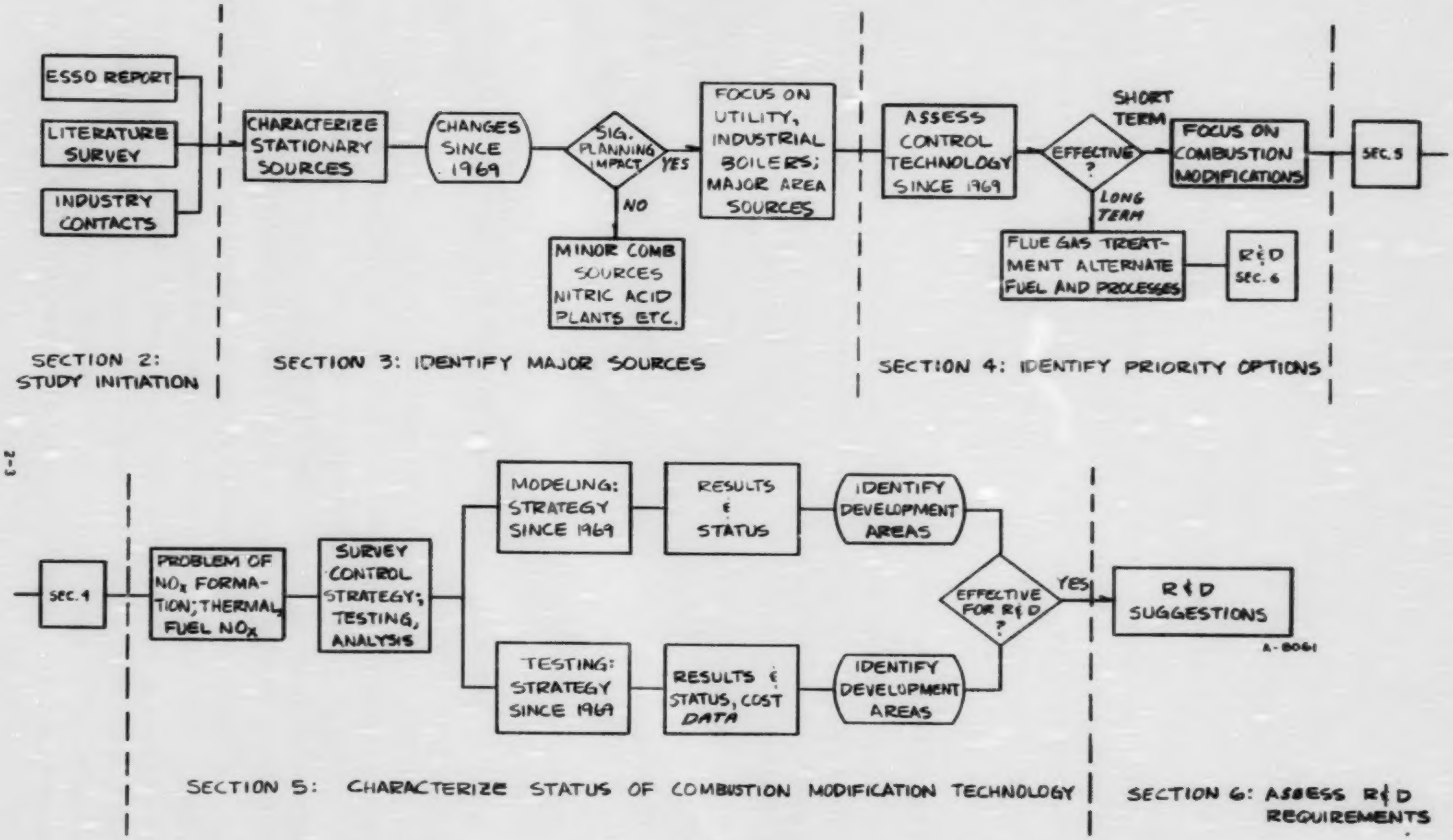

ESSO REPORT
LITERATURE SURVEY
INDUSTRY CONTACTS
CHARACTERIZE STATIONARY SOURCES
CHANGES SINCE 1969
SIG. PLANNING IMPACT
YES
NO
MINOR COMB SOURCES NITRIC ACID PLANTS ETC.
FOCUS ON UTILITY, INDUSTRIAL BOILERS; MAJOR AREA SOURCES
ASSESS CONTROL TECHNOLOGY SINCE 1969
EFFECTIVE ?
SHORT TERM
LONG TERM
FOCUS ON COMBUSTION MODIFICATIONS
SEC. 5
FLUE GAS TREATMENT ALTERNATE FUEL AND PROCESSES
R&D SEC. 6
SECTION 2: STUDY INITIATION
SECTION 3: IDENTIFY MAJOR SOURCES
SECTION 4: IDENTIFY PRIORITY OPTIONS
SEC. 4
PROBLEM OF NOx FORMATION; THERMAL, FUEL NOx
SURVEY CONTROL STRATEGY; TESTING, ANALYSIS
MODELING: STRATEGY SINCE 1969
TESTING: STRATEGY SINCE 1969
RESULTS & STATUS
RESULTS & STATUS, COST DATA
IDENTIFY DEVELOPMENT AREAS
IDENTIFY DEVELOPMENT AREAS
EFFECTIVE FOR R&D ?
YES
R&D SUGGESTIONS
A-8061
SECTION 5: CHARACTERIZE STATUS OF COMBUSTION MODIFICATION TECHNOLOGY
SECTION 6: ASSESS R&D REQUIREMENTS

Figure 2-1. Summary of the Approach to the Present Study

SECTION 3

CHARACTERIZATION OF EMISSION SOURCES

This section reviews briefly the NO_X emission inventory presented in the 1969 Esso[1] study with emphasis on recent data, significant changes, and areas of questionable data. The present study has emphasized current stationary source emissions, but includes some reflections on the projections for 1980 and 2000.

3.1 SOURCES OF NITROGEN OXIDES

No new major sources of NO_X have been discovered during the past five years. The primary sources of nitrogen oxides are many combustion processes and some noncombustion processes such as nitric acid plants. The Esso report pointed out that the NO_X formed in combustion comes from two distinct sources:

- The high temperature reaction of nitrogen and oxygen in air to form NO
- NO production from nitrogen compounds in fuels such as oil or coal (.2 - 1.5 percent of fuel weight as N)

The importance of the fuel nitrogen has received considerable study in the years since the Esso report and will be discussed in detail in Section 5.3.

The primary source of information for this section is data published in National Air Pollution Emission Trends 1940 - 1970, AP-115[2] compiled and published by the U.S. Environmental Protection Agency. Whenever possible the new data are compared directly with the Esso data. Other published sources on emission inventories for specific areas such as I.C. engines were also consulted but a detailed study as done in the Esso report was not undertaken. Areas where further information is needed will be pointed out.

Table 3-1 shows a breakdown of emission sources by type from several sources including the 1969 Esso report[1]. Some of the data in each of these other reports actually derive from the Esso study. However, any available new data which updated the Esso study were incorporated in these later surveys. More recent data (1974) from Reference 3 (NEDS data file) are included for comparison purposes and are considered very preliminary. Based on References 2

TABLE 3-1

TOTAL NO_x EMISSIONS FROM STATIONARY SOURCES (X 10^6 tons/year)

Source	EPA(3) 1974	AP-115, 1971(2)	Esso, est. 1970,(1)	Other, 1970(3,4,5)
Utility Size Boilers	5.88	4.71	3.841	3.3-6.0[c]
Recip. I. C. Engines	0.918	4.53	2.097[b]	1.95-2.3
Industrial Size Boilers	3.91		2.81	
Forest Fires (controlled)	d	.2 75	a	
Domestic And Commercial Space Heaters	0.945	0.79	1.001	
Gas Turbines	d	a	a	
Solid Waste Disposal	0.179	0.395	a	
Process Heaters	2.876	a	a	
Nitric Acid Plants	d	a	0.235	
Other Sources	d	0.338		
Total	14.7	11.038	9.986	

Notes:
- a. Included In Industrial-size Boilers
- b. Pipeline And Gas Plants Only
- c. Depending on Load Factor (High Number For Full Load)
- d. Not included in NEDS data

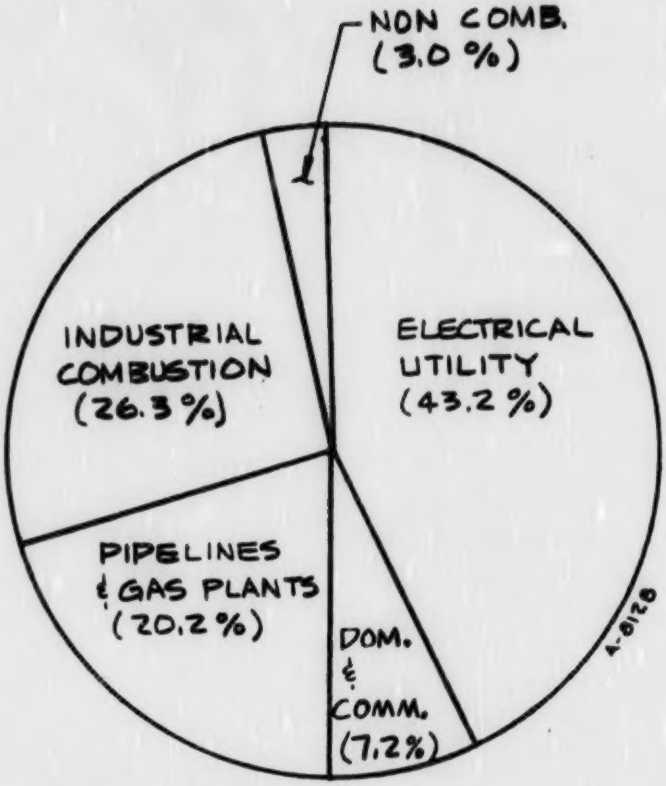
and 4, a revised pie chart was constructed as shown in Figure 3-1. This chart corresponds to the Esso pie chart (Figure 3-1 in Reference 1) but employs the AP-115 data in Table 3-1. The I.C. engine data and industrial boiler data were divided into industrial combustion and pipeline-gas plant categories on the basis of the estimated I.C. engine data from the McGowin (Shell) study[4]. Figure 3-1 indicates that the order of importance of each category has not changed. The greatest change is in the estimate of the electrical utility category, which increased from the 1970 Esso estimate of 3.84 x 10^6 tons of NO_x per year to 4.77 x 10^6 tons/year. The reason for this will be discussed under the section on large utility boilers. However, Figure 3-1 does not give a representative picture of the importance of internal combustion engines, because they are distributed between pipelines and gas plants, electrical utilities and industrial combustion. Using the figure 2.22 x 10^6 tons/year, the contribution of I.C. engines is 20.1 percent of the total stationary source emissions. This will be further discussed in Section 3.1.3 on I.C. engine emissions. A more representative pie chart can then be made which divides the sources into the following categories:

- Large utility boilers
- I.C. engines and gas turbines
- Industrial Combustion
- Residential and commercial heating
- Noncombustion sources.

This revised pie chart is shown in Figure 3-2.

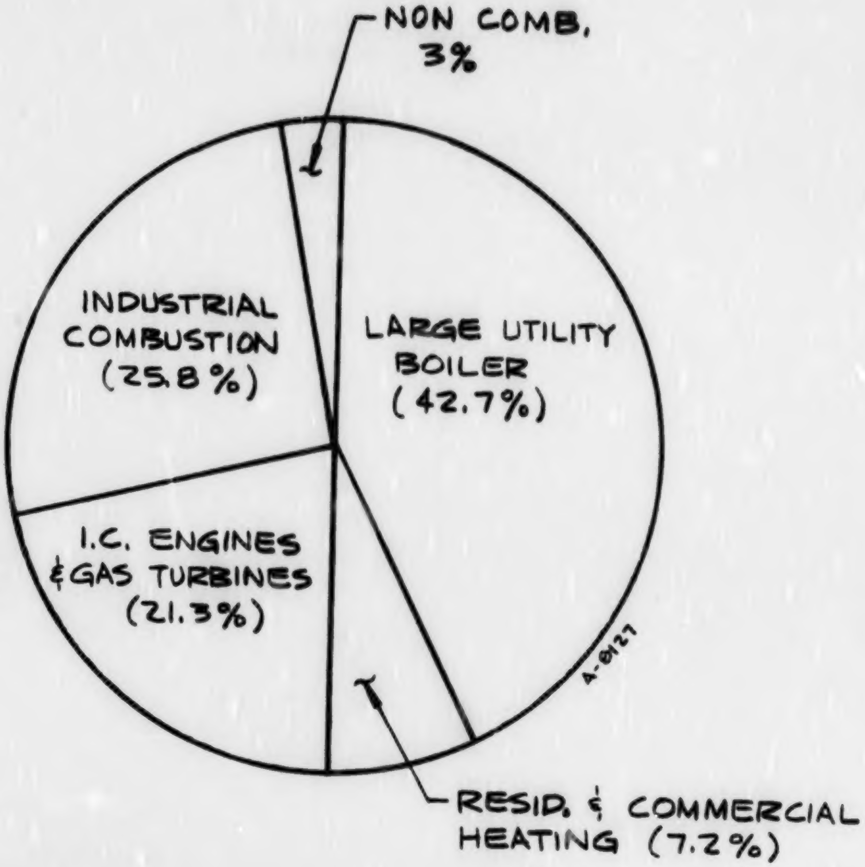
It must be remembered that data presented in total amounts does not necessarily reflect the importance of the source. Other factors such as altitude, climatology, geographical location and population density should be considered.

For the year 1971, the distribution for coal, oil and gas contributions to total NO_x production for stationary sources is shown in Table 3-2. This table shows the importance of coal to the total NO_x emissions. This importance is further amplified by considering the percentage contribution by the electric utility industry: 63.4 percent as shown in Table 3-3. This percentage will certainly increase during the next decade as some utilities convert to coal. There is also a trend to oil and away from gas. For example, according to management at two West Coast utilities their boilers were typically 70-75 percent gas-fired last year, are 40 percent gas-fired this year and will only be five percent gas-fired next year. There will also be a move in the industrial sector towards greater utilization of coal.



NO_x ESTIMATED TONS 1971	TYPE OF INSTALLATION
4,770,000	ELECTRICAL UTILITY
2,900,000	INDUSTRIAL COMBUSTION
2,240,000	PIPELINE & GAS PLANTS
790,000	DOMESTIC AND COMMERCIAL
340,000	NON COMBUSTION
11,040,000	U.S. TOTAL

Figure 3-1. Total NO_X Emitted In The U.S. From Stationary Sources, 1971.



NO_x ESTIMATED TONS, 1971	SOURCE CATEGORY
4,710,000	LARGE UTILITY BOILERS
2,350,000	I.C. ENGINES & GAS TURBINES
2,850,000	INDUSTRIAL COMBUSTION
790,000	RESIDENTIAL & COMMERCIAL HEATING
340,000	NON-COMBUSTION SOURCES
11,040,000	

Figure 3-2. Total NO_X Emitted In The U.S. From Stationary Sources, 1971. (From Data Obtained From References 2 and 4)

TABLE 3-2

1971 ESTIMATES OF NO_x EMISSIONS FROM FOSSIL-FUEL STATIONARY SOURCES

Fuel	Emissions 10^6 tons/year	Percent of* Total Stationary Sources
Gas	4.6	41.6
Oil	1.4	12.7
Coal	3.9	35.3

*Does not add to 100 percent; remainder is other combustion and non-combustible sources.

TABLE 3-3

NO_x EMISSIONS FROM FOSSIL-FUEL ELECTRICAL UTILITY UNITS

Fuel	Percent of Total NO_x Emissions From Electrical Utilities
Gas	21.6
Oil	15.0
Coal	63.4

3.2 EMISSION FACTORS

The estimates outlined above for NO_X emissions from stationary sources depend on three factors:

1. The total number of sources
2. The emission factor for each source
3. The use factor for each source

The Esso report used emission factors derived principally from the 1968 Edition of "Air Pollution Emission Factors," AP-42, with their own revision for CO boilers and catalytic cracking regenerators. The data reported in Table 3-1 from AP-115 are based on emission factors from the February, 1972 edition of AP-42, and include most of the emission factors reported in the April 1973 edition of AP-42[6].

Table 3-4 compares the emission factors from the Esso report to the latest edition of AP-42[6] and a paper by Jenkins and of McCutchen[7] for all sources which emit NO_X. Only a few sources are mentioned in the Jenkins[7] paper and these generally agree with AP-42, except for the average figure given for pulverized coal-firing of steam power plants which seems rather high. As As can be seen from the table, the AP-42[6] document goes into much greater detail than did the Esso report. When this greater detail is listed, the average Esso value is given for the category. Note also that the factors are presented next to each major heading in the units given by AP-42. (These units are not on an equivalent BTU basis.) However, most of the fuel combustion sources are grouped under the fuel type in External Combustion Sources rather than the particular industry. For example, the Esso report estimates emission from heaters, boilers, gas engines and gas turbines under gas plants, petroleum production and pipelines. Strangely, a few emission sources are missing from AP-42[6]. These sources include diesel I.C. engines in petroleum refineries, heaters, boilers and gas engines in gas plants, gas engines in pipelines, sintering and other furnaces in steel making, lime kilns and glass manufacture. Many of these may be accounted for under external combustion sources. Alternatively, sources included in AP-42[6] but not mentioned in the Esso report are LPG combustion, wood and bark combustion, tunnel kilns in brick manufacture and fiber glass manufacture. Most of these are relatively minor sources and may have been included under other headings.

TABLE 3-4

EMISSION FACTORS FOR NO_x FROM STATIONARY SOURCES

Source Category	AP-42 Rev. (Ref. 6)	Esso (Ref. 1)	EPA Paper (Ref. 7)
External Combustion Sources			
Bituminous Coal Comb. (lbs/ton)			
Pulverized			
General	18		
Wet bottom	30		
Dry bottom	18	19.98	33
Cyclone	55		
Stoker (Large)	15	19.98	
Stoker (Small)	6	8.0	
Hand Fired	3		
Anthracite Coal Comb. (lbs/ton)			
Pulverized	18	19.98	
Overfeed Stokers	6-15	19.98	
Hand Fired Units	3	8.0	
Fuel Oil (NO_x lb/10^3gal)			
Power Plant	105	103.9	105
Tangential Fired	50		
Residual	40-80	71.9	
Distillate	40-80	12.0-71.9	
Domestic	12	12	
Nat. Gas (NO_x lb/10^6ft^3)			
Power Plant	600	390	418
Tangentially Fired Power Plant	300		
Ind. Process	120-230	214	
Domestic and Com. Heating	80-120	116	
LPG Comb. (lbs/10^3gal)			
Ind. Proc. Furn-Propane	12.1	ND	
Ind. Proc. Furn-Propane	11.2		
Domes. and Com. Furn. Butane	8-12		
Domes. and Com. Furn. Propane	7-11		
Wood and Bark Comb. (lb/ton)	10	ND	

TABLE 3-4 CONTINUED

Source Category	AP-42 Rev (Ref. 6)	Esso (Ref. 1)	EPA Paper[1] (Ref. 7)
Refuse Incineration (lb/ton)			
Municipal	3	2.0	
Industrial/Commercial			
Multiple Chamber	3	2.4	
Single Chamber	2	2.4	
Trench	4	2.4	
Controlled Air	10	2.4	
Flue Fed Single Chamber	3	2.4	
Flue Fed Modified	10	2.4	
Domestic Single Chamber			
w/o Primary Burner	1	2.4	
w/ Primary Burner	2	2.4	
Pathological	3	2.4	
Auto Body Incineration (lb/car)	.1		
Conical Burner (lb/ton)			
Municipal Refuse	5	.67	
Wood Refuse	1	.67	
Open Burning (lb/ton)			
Municipal Refuse	6	10.9	
Automobile Comp.	4	10.9	
Agric. Field Burn.	2	2.0	
Landscape Refuse	2		
Wood Refuse	2		
Stationary Gas Turbines (Oil, lb/10^6Btu)	.84	.11-0.32	
(Gas, lb/10^6Btu)	.57	0.2	
Adipic Acid (lbs/ton)	12	11.6-13	
High Explosive Mfg. (lbs/ton)			
Nitration Reactors.	160	2.5-6	
Nitric Acid Conc.	1	2.5-6	
Red Water Incin.	6	2.5-6	

TABLE 3-4 CONTINUED

Source Category	AP-42 Rev. (Ref. 6)	Esso (Ref. 1)	EPA Paper[1] (Ref. 7)
Low Explosive Mfg. (lbs/ton)			
Nitrocellulose Reactor Pot.	12	2.5-6	
Nitrocellulose Sulfuric Acid Conc.	29	2.5-6	
Nitric Acid Mfg. (lb/ton acid)			
Weak Acid Uncontrolled	50-55	57	43
Catalytic Comb. N.G.fired	2-7		
Catalytic Comb. H_2 fired	0-1.5		
Catalytic Comb. Mixture	.8-1.1		
High Strength Acid	.2-5.0		
Coffee Roasting (lbs/ton)	0.1	Neg.	
Nitrate Fertilizers (lbs/ton)		Neg.	
w/Granulator	0.9		
Dryers & Coolers	3.0		
Sugar Cane Processing			
Field Burning (lb/acre burned)	30		
Blast Furnace (lbs/ton of pig iron)	ND	.397	
Coke Manufacture (lb/ton)			
Charging	.03	.065	
Coking Cycle	.01	.065	
Cupola Furnace	ND	.11	
Steel Foundries (lb/ton)			
Electric Arc	.2	ND	
Open Hearth	.01	2.45	
Other Furnaces in Steel Making	ND	.086	
Sintering	ND	1.04	
Brick Manufacture (lbs/ton)			
Tunnel Kilns (lb/ton)			
Gas Fired	.15	ND	
Oil Fired	1.1	ND	
Coal Fired	0.9	ND	

TABLE 3-4 CONTINUED

Source Category	AP-42 Rev. (Ref. 6)	Esso (Ref. 1)	EPA Paper[1] (Ref. 7)
Periodic Kilns			
Gas Fired	0.42	1.68	
Oil Fired	1.7	3.84	
Coal Fired	1.4	4.128	
Cement Mfg. Kilns (lbs/ton)	2.6	1.53-3.69	
Lime Kilns	ND	1.152-2.74	
Fiber Glass Mfg. (lb/ton)			
Textile Products			
Glass Furnace			
Regenerative	9.2	ND	
Recuperative	29.2	ND	
Curing Oven	2.6	ND	
Wool Products (lbs/ton)			
Glass Furnace			
Regenerative	5.0	ND	
Recuperative	1.7	ND	
Electric	0.27	ND	
Curing Oven	1.1	ND	
Cooling	0.2	ND	
Petroleum Refineries			
Boilers & Process Heaters			
Oil Fired (lb/10^3bbl)	2900.	2800	
Gas Fired (lb/10^3ft^3)	.023	.21	
Fluid Cat-Cracking Units (lb/10^3bbl fresh feed)	71	4.2	
	(37.1-145.0)	4.2	
Moving Bed Cat Cracking (lb/10^3bbl fresh feed)	5	4.2	
Compressor I.C. Engines (lb/10^3ft^{3}gas burned)	0.9	.77-4.35	
I.C. Engines-Diesel (lbs/10^3bbl)	ND	.0093	
Gas Turbines (lb/10^3ft^{3}gas burned)	ND	.2	

TABLE 3-4 CONCLUDED

Source Category	AP-42 Rev. (Ref. 6)	Esso (Ref. 1)	EPA Paper[1] (Ref. 7)
Gas Plants (lbs/10^3ft^{3}gas burned)			
Heaters & Boilers	ND	.19	
Gas Engines	ND	4.3	
Gas Turbines	ND	.2	
Pipelines (lbs/10^3ft^{3}gas burned)			
Gas Engines	ND	7.3	
Gas Turbines	ND	0.2	
Glass Mfg. (lb/10^6Btu)	ND*	.87	

*Included most likely as external combustion sources

3.3 PROSPECTS OF NO_X EMISSIONS

The data presented in the previous sections are continuously being refined and updated as more detailed studies of each source and data from field tests become available. This section discusses these changes for each major stationary source as well as the factors which will influence the emissions in the future. These factors include

- Changes in energy demand
- Fuel switching
- Impact of NO_X controls.

3.3.1 Large Utility Boilers

3.3.1.1 Emissions Factor Adjustments and Refinements

The greatest change in the emission factors for large utility boilers has been for gas-fired plants. Esso reported 390 lbs $NO_X/10^6 ft^2$ of gas and AP-42 reported 600 lbs $NO_X/10^6 ft^3$ of gas. Between the gas emission factor change and more detailed factors for each type of coal-fired unit, the overall emissions have increased from the 1970 Esso estimate of 3.84×10^6 tons/year to 4.71×10^6 tons per year.

It should be noted that these estimates do not include the effect of emission controls. However, other than in West Coast utilities and new plants across the U.S., there has been very little application of NO_X controls to date, especially on oil- and coal-fired plants. The gas- or oil-fired utilities on the West Coast have seen reductions in NO_X by a factor of 2.

3.3.1.2 Relative Significance of Different Fuels

Converting the revised emission factors in Table 3-4 to an equivalent BTU basis gives a better indication of the relative importance of each fuel, as shown in Table 3-5.

TABLE 3-5

COMPARISON BETWEEN COAL, OIL AND GAS ON EQUIVALENT BTU BASIS; ELECTRIC POWER GENERATION (lbs $NO_x/10^9$ Btu)

Natural Gas (1046 BTU/ft^3)	574
Fuel Oil (149,966 BTU/gal)	700
Coal (avg) (11,867 BTU/lb)	758

Thus, for any given plant there should be an 8 percent increase in NO_X when converting from oil to coal and a 30 percent increase when going from gas to oil provided the load remains constant. If the unit was designed to operate on dual fuels the load will most likely not be lowered. However, if the boiler was not initially designed to operate on a different fuel, converting from gas to oil to coal will generally reduce load.

3.3.1.3 Projections for the Future

Projections for the future are speculative at best. In the very short term we see many utilities decreasing their load from 5 to 15 percent to meet the "energy crisis" in fuel supply. This reduction in load will yield a corresponding reduction in NO_X. The duration of this situation is even more speculative. However, longer term projection as shown in Figure 3-3[8] indicate continuing increase in coal, oil, and gas generating capacity with a substantial increase in nuclear capacity.

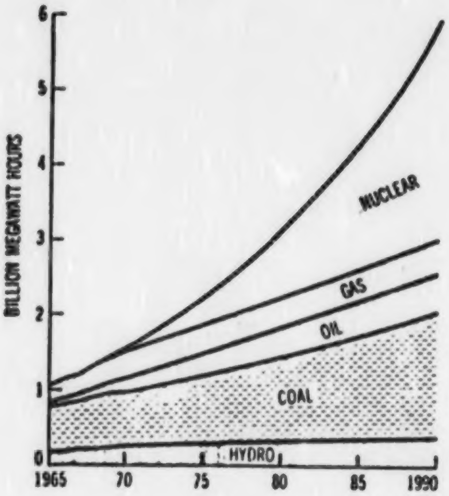


Figure 3-3. Estimated Annual Electric Utility Generation By Primary Energy Sources

Projections such as this assume that the fuel will be available and that the generating stations will be built to meet the demand. In fact, the future use of fossil fuels is difficult to project. Figure 3-4 shows an estimated breakdown of the total U.S. energy demand by each domestic energy source and the required imports to meet the demand. This particular estimate shows that by 1975, 15.3 percent of the energy could be imported, chiefly as petroleum for

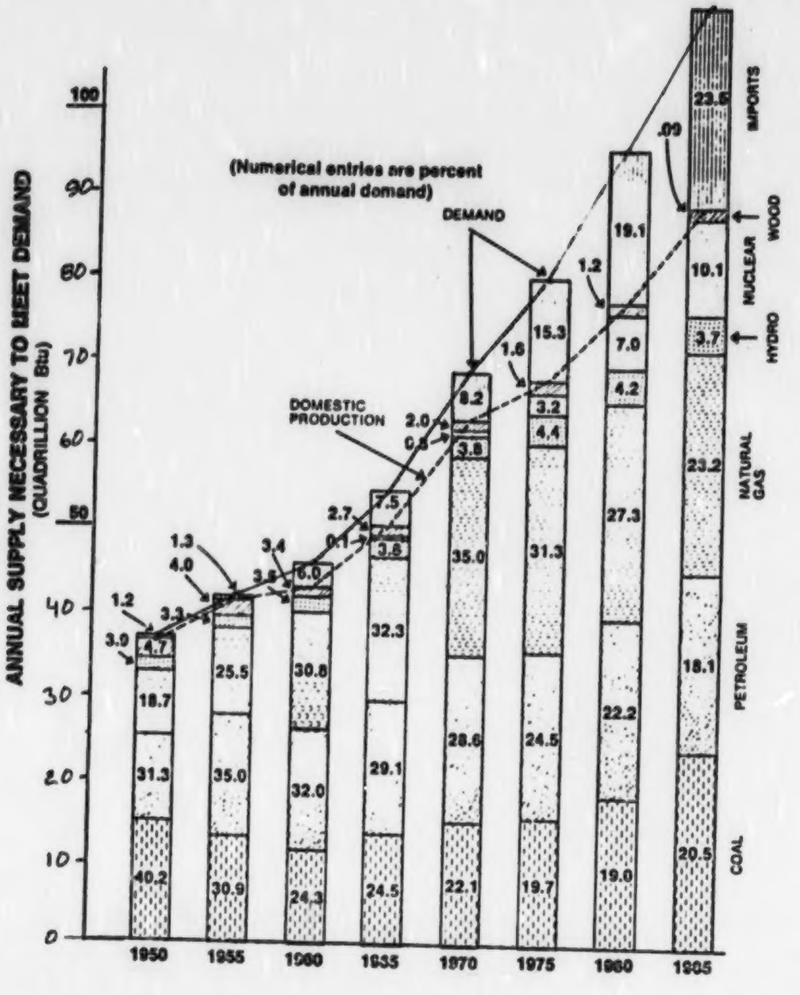


Figure 3-4. Trends Of Sources Of U.S. Energy Supply; 1950-1970(Actual)/1975-1985(Projected)

which the availability and price depend on the international political situation. Even with adequate domestic and imported oil and gas we have seen shortfalls in getting the energy to the public due to delays in increasing refinery capacity because of the relative attractiveness of investment abroad and because of environmental considerations.

With the recent emphasis toward becoming energy independent there is certainly going to be an effort made to utilize the 3.2 trillion tons of potential coal supplies in the U.S. In order to meet the short term energy gap the sulfur regulations on coal may have to be relaxed and there will probably be a greater utilization of coal than indicated on Figures 3-3 or 3-4. In the longer term, however, there also may be a number of problems in meeting the demand for coal for direct combustion and, eventually, for coal gasification and liquifaction. These problems include:

- Mining capacity
  - Trained miners
  - Availability of mining equipment
  - Process water
- Large investment requirements
- Transportation of the coal to the utility
- Environmental Considerations
  - Use of low sulfur coal
  - Strip mining overburden
  - Scarred landscape
  - Water pollution from acid mine drainage
  - Underground mining resulting in land subsidence with time
  - Coal preparation waste; slag heaps

Thus to what extent and how fast the coal actually gets utilized is also very speculative, and, as listed above, similar uncertainties make it difficult to project oil usage.

We see the following non-quantified trends in the electric utility industry:

- In the short term 1974 - 1980, increased use of direct combustion of coal
- Switching from gas to oil where feasible

- **Accelerated use of nuclear energy**
- **Emphasis on "clean fuels" for the 1980 - 2000 period**
  - **Synthetic gas from coal**
  - **Synthetic oil from coal**
  - **Shale oil**
  - **Fluidized bed combustion of coal**
  - **Combined cycle schemes using low BTU synthetic gas, synthetic oil, or fluidized bed combustion of coal.**

It is not presently possible to project the potential impact of these fuels switching trends on total NO_x emissions except to observe that at least in the short term the NO_x problem should intensify. The trends cannot be quantified with any certainty, and in any case there is presently no information with regards to NO_x emissions from combustion of synthetic gas or oil, shale oil or the use of low BTU gas in combined cycle schemes. Some preliminary information is available on fluidized beds combustion of coal. This will be discussed in Section 4.1.3.1.

3.3.2 Domestic and Commercial Sources

Additional data on emissions factors for home heaters and commercial boilers are shown in Table 3-6 from a report by Barrett[10]. This data was developed under EPA and American Petroleum Institute sponsorship and updates the AP-42 document[6]. The recommended emission factors are 67 percent higher for oil-fired residential units and approximately the same for commercial boilers. Further studies by Rocketdyne under EPA contract will help to define the level of controllable emissions by optimizing the combustor.

The trends predicted in the Esso study are still valid, although the levels may be higher due to recent emission factors. However, if the energy shortfall continues the following events may occur:

- Reduced emissions due to reduced load (cooler homes)
- Some switching back to coal-fired home heating systems
- In the longer term switching to electrical heat (provided the power plants can meet the demand)
- Increases in efficiency of the home heater system and greater housing insulation to reduce the total load and thus emissions
- By the year 2000, some homes using solar heating.

TABLE 3-6

NO_x EMISSION FACTORS FOR RESIDENTIAL AND COMMERCIAL OIL-FIRED HEATERS AND BOILERS (lbs/10^3 gal)[6,10]

Residential Heaters	
Battelle-Suggested Emission Factor	20.0
AP-42 Emission Factor	12.0
Commercial Boilers	
Battelle-Suggested Emission Factors	
No. 2 Oil	$20 + 78N^{0.6}$
No. 4	$20 + 85N^{0.6}$
No. 5	$20 + 87N^{0.6}$
No. 6	$20 + 89N^{0.6}$
LSR (Low Sulfur Resid.)	$20 + 84N^{0.6}$
AP-42 Emission Factors	
Distillate	20 - 40
Residual	20 - 40

where: N = multiplication factor equal to percent nitrogen in fuel oil. Typical values are:

GRADE	N
2	.01
4	.2
5	.3
6	.4
LSR	.2

3.3.3 Stationary I.C. Engines and Gas Turbines

Total emissions from stationary I.C. engines and gas turbines are probably the least-well documented and are now believed to represent from 20 to 30 percent of the stationary source emissions (Table 3-1). Table 3-7 gives an indication of the problem by showing the total estimated horsepower and emissions of I.C. engines in the U.S. by several authors including Esso. As can be seen in the table, estimates of installed horsepower and of the total emissions vary by a factor of two and do not always correspond. It should be noted, however, that the Esso emissions only include the gas pipeline and oil and gas production sources. Preliminary data generated by Aerospace[5] under EPA contract agree fairly closely with the Shell[4] estimate done under an earlier EPA contract. Table 3-8 summarizes the emission factors used for each engine type by Esso and Shell. In general, the emission factors do not differ greatly, although the Shell data are usually higher. It should be remembered, however, that a significant percentage of these engines (gas pipeline and oil and gas production engines) are located in non-urban areas and that the residence time of NO or NO_2 in the atmosphere is only a few days[1].

Installed horsepower and estimated emissions from stationary gas turbine engines are presented in Table 3-9. There is considerable discrepancy between the Esso numbers and the Shell report; however, the total emissions amount to only 1 percent of the total stationary source emissions. The electric power generating turbines chiefly represent standby and peaking units (although an increasing number are coming on as base load units) but are important since they occur in urban areas. When on line they can represent a sizeable point source of emisssions. The Aerospace study[5] will also be taking a better look at these emission sources.

We see the following trends with regard to I.C. engines and gas turbines.

- Significant increase in the use of gas turbines for gas pipelines, oil field application and peaking and standby units for electrical power generation
- Continued increase in I.C. engine applications but many applications going to gas turbines for greater reliability (unless fuel costs becomes prohibitive)
- Increased use of gas turbines for combined cycle plants.

TABLE 3-7

TOTAL INSTALLED HORSEPOWER AND EMISSION FROM RECIPROCATING I.C. ENGINES

Engine Type	Installed Horsepower		
	Esso[1]	Shell[4,a]	Aerospace[5]
	10^6 hp	10^6 hp	10^6 hp
Gas Engines			
Oil & Gas Transmission Lines		11.28	
Utility Electric Power Generators		3.80	
Agricultural Wells	53.0	0.00	
Oil & Gas Production	(total)	7.00	
Commerical/Institutional/Industrial		0.23	
Municipal Water & Sewer Treatment		0.46	
Oil Fueled Engine			
Diesel - Precombustion Supercharged	0.7		
Diesel - Precombustion Nonsupercharged	0.1	11.86	
Diesel - Direct Injection Supercharged	14.0		
Diesel - Direct Injection Nonsupercharged	2.0		
Total Horsepower	69.8	37.74	
Engine Application	NO_x Generation by I.C. Engines (tons/year)		
	10^6 ton/yr	10^6 ton/yr	10^6 ton/yr
Peaking Power	?	0.062	0.065-5[b]
Gas Pipeline	1.596	0.930	?
Oil & Gas Production	0.730	0.832	?
Agricultural Wells	?	0.319	?
Standby	?	0.0	?
Other	?	0.093	?
Total Emissions	(2.326)	2.236	1.95

[a] Includes dual fuel engines.

[b] Includes peaking power and standby from normal load to full load capacity.

TABLE 3-8

NO_x EMISSION FACTORS FOR RECIPROCATING I.C. ENGINES (gm.NO_x/Bhp-Hr)

Engine	Supercharged?	Esso[1]	Shell[4]	AP-42[6]
Diesel, Precombustion Chamber	Yes	3.36	5.3	3.8-22.7
Diesel, Precombustion Chamber		1.37-1.95	5.9	
Diesel, Direct Injection	Yes	11.75	13.8	
Diesel, Direct Injection		4.29-9.17	11.2	
Natural Gas (Otto)	Yes	25.4	12.5	
Natural Gas (Otto)		12.7	11.8	

TABLE 3-9

TOTAL ESTIMATED HORSEPOWER & EMISSIONS FROM GAS TURBINES

	Horsepower (10^6hp)	Emissions (tons/yr)
Esso		
Turbojet - Oil	5	?
Turbojet - Gas	46	?
Esso Total	51	21,000[a]
Shell		
Electric Power Gen.	30.44	62,920
Oil and Gas Pipelines	3.52	39,800
Natural Gas Processing Plants	1.53	28,130
Shell Total	35.49	130,200

[a] Includes only oil and gas piplines (19,000 tons/year) and natural gas processing plants (2,000 tons/year)

An EPA grant (No. R80227D with Aerospace Corporation) will provide further data on I.C. engines and will recommend an R&D program for I.C. engines as a source of NO_x[5].

3.3.4 Industrial Combustion Sources

Industrial combustion sources include industrial boilers and process heaters, total energy systems, incineration and other burning, metallurgical processes, kilns for cement, limestone, and ceramics, and glass manufacture. As can be seen by studying Table 3-4, many of the emission factors have been further updated and more finely divided but the overall effect on total emission from industrial processes has not changed much from the Esso prediction. Except for industrial package boilers, which contribute a significant percentage of NO_x to this area source, very little testing has been done to date. Again, the Aerospace study[5] will provide additional estimates on NO_x emissions from these industrial area sources. Further testing of these area sources is planned by the Combustion Research Section, Control System Laboratory of EPA. Trends for the future include:

- Steady growth in the number of plants
- Perhaps some fuel switching to oil or coal depending on
    - Environmental laws
    - Price availability of fuel
- Higher preheat and O_2 enrichment for energy efficiency.

3.3.5 Non-Combustion Sources

As has been mentioned in numerous references the primary noncombustion source of NO_x emissions is in the manufacture of nitric acid. Emission factors are now more detailed but the total emissions are approximately as predicted by Esso.

Discussions with EPA personnel[11] indicate that commercial and economical control schemes have been developed (e.g., Union Carbide-Molecular Sieve scheme) and could soon reduce the emission to 10 - 25 ppm from an uncontrolled value of around 200 ppm.

3.4 NO_x EMISSION TRENDS

The trend of NO_x emission from stationary sources from 1950 to 1971 from from AP-115[2] is illustrated in Figure 3-5. Also shown in Figure 3-5 is the Esso predicted curve from 1950 to the year 2000. As can be seen on the curve NO_x levels have already reached the 1975 level predicted by Esso. The slight downward trend in 1971 is due to revised emission factors and implementation

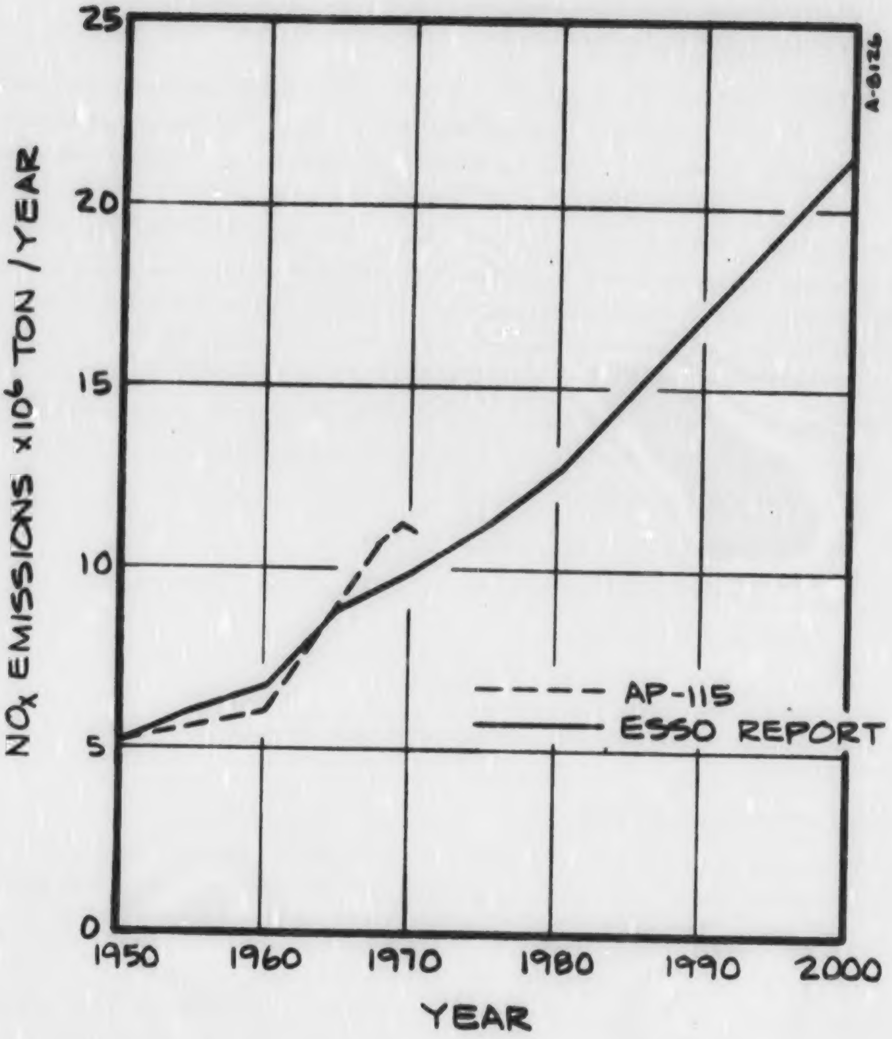


Figure 3-5. Stationary Source NO_X Emission Trends

of NO_X controls on the West Coast. If further controls are implemented both on stationary and mobile sources the potential trend may be as illustrated in Figure 3-6[12]. This figure shows the contribution for controlled and uncontrolled emission of each fuel type for total NO_X emissions. However, Figure 3-6 does not reflect the impact of fuel switching. Additional studies to predict trends which incorporate recent developments are necessary.

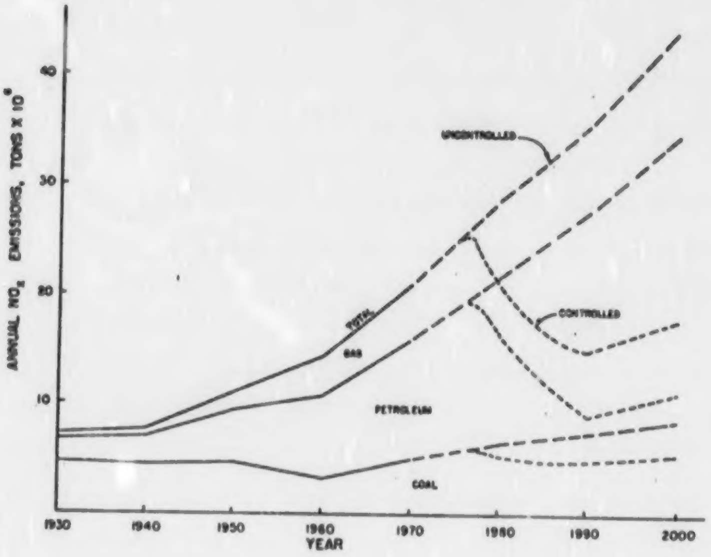


Figure 3-6. Stationary And Mobile NO_X Emission Trends

3.5 SUMMARY

Recent data have not substantially altered the NO_X contributions reported by the Esso study. However, it must be remembered that a straight ranking of amounts of emissions does not consider the importance of factors such as altitudes, dispersion, climate, geographical location, etc. These other factors are particularly important when considering I.C. engines. New data brought out in this section include the following:

- NO_X emissions from steam electric generating are higher than predicted by the Esso report for 1970 - 1971.
- NO_X emission quantities from I.C. engines and gas turbines is not well defined, but may contribute 20-30 percent of the total.
- The effects of NO_X emission from I.C. engines on the overall air quality is unknown.

- Emission factors have been updated with recent data and are now more detailed in all categories.
- Emission factors for oil fired residential heaters are 67 percent higher than reported by Esso.
- There is very little field data on industrial combustion sources.
- NO_X controls have been incorporated on West Coast electrical power plants.

Trends for the future include the following:

- There will be a significant increase in the utilization of coal and oil in power generation, leading to an intensified NO_X problem which is impossible to quantify at this time.
- Industrial area sources may be switching to oil or coal if the energy shortage continues, leading to an intensified NO_X problem which is impossible to quantify at this time.
- Home heating systems will become more efficient if the cost of fuel rises and we may see a smaller increase in NO_X emission as a result.
- The degree to which NO_X controls will be implemented in other areas depends on local and federal regulations, such as the New Source Performance Standards (NSPS), amount of fuel switching and further developments for oil and coal.

Recommendations for future work to more clearly define the status of emissions include:

- Those sources which seem to be missing from AP-42[6] should be investigated to determine if they appear in some other category.
- A better assessment of the relative importance of each source should be made.
- Additional field data is necessary for industrial combustion sources, I.C. engines and gas turbines.
- A study is necessary to determine a reasonable implementation plan for NO_X controls on stationary sources and to determine the impact on NO_X emissions.

SECTION 4

SURVEY OF OPTIONS FOR NO_x CONTROL

The preceding section surveyed NO_x emission sources with a view toward identifying changes since the publication of the Esso report which would alter the focus of control program planning. This section gives a corresponding survey of candidate NO_x control techniques as applicable to the sources characterized in Section 3. The objective here is to identify the attractive short and long-term options for NO_x control through evaluation of developments and user experiences subsequent to the Esso study.

Short term options for NO_x control necessarily involve retrofit modifications of existing units. Following the Esso study, there has been extensive testing and application of retrofit concepts. Long-range options include incorporation of unit modifications into optimized design for new units as well as use of alternate processes. The selection of alternate processes is usually motivated by process economics for which pollution control devices are an important tradeoff. Increasing recognition of projected gas and oil fuel shortages, subsequent to the Esso study, has given impetus to the design and evaluation of alternate processes, particularly for power generation.

The criteria for evaluation of short and long-range options include:

- Cost effectiveness
- Availability (short term options)
- Low-risk development (long-range options)
- Impact on unit efficiency
- Impact on operational difficulties
- Overall environmental impact due to process control
- Compatibility with projected fuel allocations

Options for NO_x control from combustion sources are evaluated in Section 4.1; non-combustion sources are treated in Section 4.2.

4.1 COMBUSTION SOURCES

Combustion generated NO_x results from the thermal fixation of atmospheric nitrogen in the combustion air, or from conversion of bound nitrogen in the fuel. Thermal NO_x can be formed in the oxidation of virtually any fuel in the presence of air, while fuel NO_x results primarily with firing of coal and the heavier residual and crude oils. From a systems standpoint, the control of NO_x emissions can be approached by four strategies:

- Modification of operating conditions in existing units to alleviate conditions favorable to NO_x formation;
- Modification of fuel by denitrification, use of additives, or substitution with a low NO_x forming fuel;
- Treatment of flue or exhaust gas for NO_x removal;
- Use of alternate low NO_x processes in new units.

Control techniques in these categories are reviewed below.

4.1.1 Modification of Existing Units

The Esso report concluded, on the basis of the limited data available, that retrofit modification of existing units was the most promising approach to the reduction of NO_x emissions. In the intervening period, the feasibility and some limitations of this strategy have been demonstrated, particularly for utility boilers. Evaluations of the combustion modification strategy are given below for boilers, stationary engines, and other combustion sources.

4.1.1.1 Utility and Industrial Boilers

Utility boilers are the largest single stationary source of NO_x emissions and are also "point sources" in the sense of having a very high ratio of total emissions to the number of units. These units have accordingly been the most extensively modified as part of the NO_x control program. Industrial boilers are usually regarded as an "area source," with a low ratio of emissions to number of units, and have been accorded a lower priority in the NO_x control program. The technology for modification of these units is currently under development utilizing, where possible, experience derived from the modification of utility boilers.

The most significant achievement in NO_x control has been with gas fired units. Typically, reductions from uncontrolled levels of 400-1000 ppm to a controlled level on the order of 200 ppm have been attained through reduction of the peak flame temperature and through creation of fuel-rich conditions in the primary flame zone. The cost-effective techniques favored to produce these changes in operational conditions are off-stoichiometric combustion by biased burner firing, reduction of excess air, and, less frequently, recirculation of flue gas into the primary combustion air. Figure 4-1 shows results from the modification of a 750 MW horizontally-opposed, face-fired unit[13].

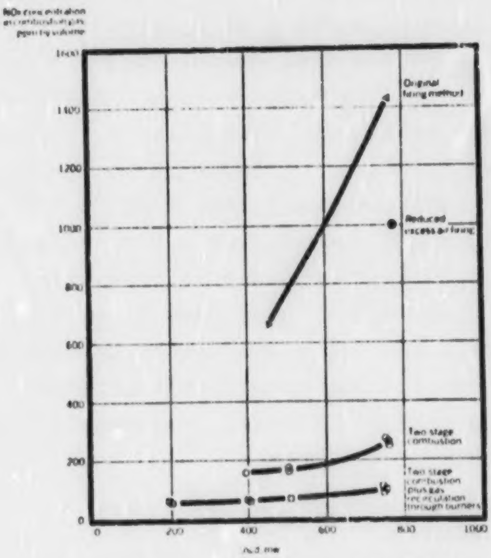


Figure 4-1a. Effects of Nitric Oxide Control Methods (Natural Gas Fuel)

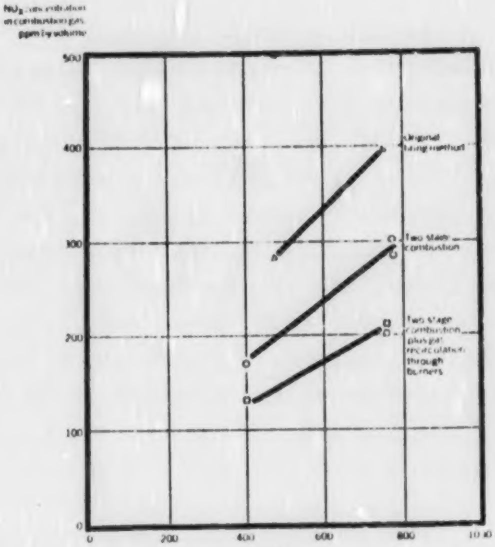


Figure 4-1b. Effects of Nitric Oxide Control Methods (Oil Fuel)

These results also show that load reduction is effective for NO_x control; use of this strategy is usually not favored, however, because of ecomonic considerations.

With oil and coal firing, the combustion modification strategy must confront the formative mechanisms for both thermal and fuel NO_x. Since the Esso study, the validity of combustion modifications for NO_x reduction in oil-fired units has been demonstrated. Operational guidelines for coal-fired units are currently being formulated through field testing. In general, tests indicate that emission reductions for oil and coal-fired units are less than for gas-fired units with comparable combustion modification. The most effective scheme is a combination of low excess air and off-stoichiometric firing. Indications are that the effectiveness of flue gas recirculation decreases as the quantity of bound nitrogen in the fuel increases.

Figure 4-1b shows typical reductions from modification with oil firing for the same unit as the gas fired results in Figure 4-1a. Although the modifications yielded significant emission reduction, the percent reduction with each modification is less with oil than with gas. The minimum emission level achieved through the combined modifications was higher for oil than for gas. For the firing of coal, the minimum emission level demonstrated through combustion modification is higher than for either oil or gas. This pattern has significant impact in view of the increasing conversion of utility boilers to coal firing.

Although combustion modifications remain as the most attractive option for reduction of uncontrolled emissions, there are limitations to the effectiveness of this strategy for attaining very low emission levels. Limitations arise through operational difficulties such as burner instability or corrosion, emission of other pollutants such as smoke or CO, or a reduction in thermal efficiency. For this situation, the diminishing returns from further application of combustion modification would eventually justify, on a cost effectiveness basis, use of alternate techniques, such as flue gas treatment, for a backup control technique. Most modifications to date have been done without optimizing burners for combustion modifications. Improved burner designs combined with other modification techniques should show significant effects. Further results and evaluations for the combustion modification strategy are given in Section 5.

4.1.1.2 Gas Turbine and Reciprocating Engines

The NO_x control technology for stationary I.C. engines and gas turbines has developed from the automotive industry and aircraft industry respectively. Each of these sources will be discussed separately.

Reciprocating I.C. Engines

Equipment modification control techniques for existing units include the following:

- Modification of operating conditions
    - Speed
    - Torque/Load
    - Ignition timing
    - Fuel injection timing
    - Air temperature
    - Air pressure
    - Exhaust back pressure
- Exhaust gas recirculation
- Water injection

- Valve timing
- Compression ratio

All of the operating conditions can be changed in a direction to decrease NO_x emission. The modifications mechanism in reducing NO_x is in general to lower the flame temperature, decrease the availability of oxygen or to shorten the equivalent residence time. The general effect of these changes when moving to lower NO_x is to increase fuel consumption, increase emission of CO,H/C or smoke or to reduce performance. The relationship between each of these operating parameters will be discussed in Section 5. Exhaust gas recirculation and water injection have also been shown to be an effective method of NO_x reduction in I.C. engines. Side effects include increased fuel consumption and possibly less engine reliability and life. Increased valve overlap has similar effects as E.G.R. and could be implemented by simply changing the camshaft. Considerable data has been developed in each of these areas since the Esso study and will be reported in Section 5. At this time the most viable technique in the short term for both diesel and spark ignition gas engines is water injection.

Gas Turbines

Potential equipment modification control schemes include the following:

- Minor combustor can modification
- Reduced turbine inlet temperature by reducing power
- Exhaust gas recirculation
- Water injection

Although considerable data has been developed in the last few years for each of these schemes no single method has been ruled out or achieved the desired reduction. All are still undergoing investigation. In the short term some electric utilities are trying water injection but feel the ultimate solution will be through new combustor can development which will provide a replacement can. Recent developments will be discussed in Section 5.

4.1.1.3 Other Combustion Sources

Other combustor sources for NO_x include the following industrial sources.

- Metallurgical furnaces
- Cement, lime and ceramic kiln
- Glass manufacture
- Incineration and waste disposal
- Residential and commercial space heating

No specific NO_x control technology was found in the recent literature for any of these areas. The Battelle study[10] surveyed emission from space heaters and showed the effect of excess air on NO_x emissions and other pollutants for 33 existing oil-fired units (decreasing excess air lowers NO_x emissions). Data reported in a paper by Howekamp[14] showed similar results for five "combustion improving" oil-fired burner heads for use in residential heaters. The Howekamp data showed a peak NO_x emission at a stoichiometric air-fuel ratio around 1.6 and decreasing on either side of this point. None of these devices substantially changed NO_x emissions. Rocketdyne[15], currently under contract with EPA to develop an "optimum" home heater size burner, have developed a low excess air burner with 30 percent to 50 percent reduction in NO_x[15]. This design is currently undergoing long term cyclic testing. Aerospace Corporation[15] under EPA contract is currently working on technology assessment of the other combustion sources but no data have been reported. This will be followed by a field study to better determine the emission factors and current range of control. Thus, the general discussion presented in the Esso report for NO_x control for each of these sources is still valid.

4.1.2 Fuel Modification

Fuel Switching

For a NO_x control strategy, natural gas firing is attractive because, as discussed in Section 4.1.1.1, the effectiveness of combustion modification is better than for oil or coal firing, particularly with high levels of fuel nitrogen, and the optimum emission level is thought to be lowest for gas. Despite the superior cost effectiveness of NO_x control in gas-firing, the economic considerations in fuel selection are currently dominated by the petroleum supply deficiency, and the trend, as stated in Section 3, is toward coal-fired units. On a short-term basis, fuel switching to natural gas or low nitrogen oil is not a promising option.

A promising long range option is the use of clean synthetic fuels derived from coal[16,17]. Candidate fuels include low Btu gas (100-300 Btu/scf), intermediate Btu gas (300-500 Btu/scf) and synthetic oil. Process and economic evaluations for the use of these fuels in power generation are being made by the EPA, The Office of Coal Research, The Bureau of Mines, and the Electric Power Research Institute. Two options considered likely are firing of low-Btu gas in a conventional boiler, and the use of low Btu gas in a combined gas and steam turbine power generation cycle. For both systems, economics favor placement of both the gasifier and power generation cycle at the minehead, so the most extensive use of these options would be for new units to replace fossil fuel units starting around 1980.

The NO_x emissions from low Btu gas-fired units are expected to be low due to reduced flame temperatures characteristic of the lower heating value fuels. The effects on NO_x emission of the molecular nitrogen and the intermediate fuel nitrogen compounds, such as ammonia, in the low Btu gas have not been determined and require study.

As a NO_x control strategy, the use of synthetic fuels is promising in the long term but will not remove the burden from combustion modifications in the short term.

Fuel Additives

In principle, use of fuel additives could reduce NO_x emissions through one or a combination of the following effects[18,19,20]:

- Reduction of flame temperature through increased thermal radiation or dilution
- Catalytic reduction or decomposition of NO to N_2
- Reduction of local concentrations of atomic oxygen

Martin *et al*. tested 206 fuel additives in an oil-fired experimental furnace[18], and four additives in an oil-fired package boiler[19]. None of the additives tested reduced NO emissions but some additives containing nitrogen increased NO formation.

Shaw[20] tested seventy additives in a gas turbine combustor and found that only metallic compounds promoting the catalytic decompostion of NO to N_2 had a significant effect on NO emissions. From 15 percent - 30 percent reductions in NO_x were achieved with addition of 0.5 percent by weight of iron, cobalt, manganese, and copper compounds. The use of this technique for NO_x control is not attractive, however, due to increased cost, serious operational difficulties, and the presence of the additives, as a pollutant, in the exhaust gas.

An indirect reduction of NO_x could result from use of additive metals intended to prevent boiler tube fouling[21,22]. The excess air level in oil-fired boilers is frequently set sufficiently high to prevent tube fouling. Use of additives could allow use of a lower level of excess air which in turn would reduce NO_x emissions. The emission reduction from this strategy is quite limited and the cost effectiveness is likely to be poor.

Fuel Denitrification

For firing of heavy oils and coal, NO_x emission reduction could be achieved through removal of the chemically bound nitrogen from the fuel. No effective denitrification process has been demonstrated and progress is hindered somewhat by uncertainty as to the types of compounds comprising the fuel nitrogen. There is some speculation that the desulfurization treatment has a side benefit of reducing fuel nitrogen, but this has not been verified.

In view of the attractive cost effectiveness of combustion modification, and the projected conversion to clean synthetic fuels, denitrification does not appear to be a promising long range options for NO_x control.

4.1.3 Alternate Processes for New Units

For new units, the combustion control technology derived from retrofit of existing units can be incorporated, together with new concepts not applicable for retrofit, into designs optimized for NO_x control. The flexibility of this approach yields potentially lower costs and higher effectiveness relative to the retrofit of existing units. Alternatively, the economic factors involved in projected fuel conversions and emission control for use of high sulfur fuels may dictate selection of an alternate combustion process.

4.1.3.1 Utility and Industrial Boilers

Optimized Design

Retrofit concepts applicable to new design include: optimized burner placement and biasing for off-stoichiometric combustion, flue gas recirculation, and overfire air ports for two-stage combustion. New design concepts include optimized burner design, particularly for suppression of fuel NO_x, and larger furnace volume. Ideally, the combustion chamber design should be tailored to the flame shape found from burner optimization studies to yield the lowest NO_x emission. One design which approaches this concept is the tangentially fired boiler. The low heat release rate and slow mixing of oxygen with the fuel in tangential firing yields relatively low rates of formation of both thermal NO_x and fuel NO_x.

With implementation of the above techniques, boiler manufacturers have been able to meet the EPA New Source Performance Standards (NSPS):

Coal	0.7 lb $NO_2/10^6$Btu
Oil	0.3 lb $NO_2/10^6$Btu
Gas	0.2 lb $NO_2/10^6$Btu

For units going into service beyond 1980, this approach to NO_x control may be replaced by use of alternate processes, discussed below.

Alternate Processes

In the period beyond the year 2000 it is projected that the U. S. energy requirements will largely be met by a combination of fission and fusion reactors and solar energy conversion supplemented by fossil fuels, MHD, and geothermal power[23]. In the interim, fossil fuels will remain the major energy source with increasing use being made of low-grade high-sulfur coals. Two promising concepts for the use of these coals in the clean and efficient production of energy are use of synthetic fuels derived from coal, discussed in Section 4.1.2, and fluidized bed combustion.

Suggested advantages of fluidized bed combustion compared to conventional boilers are[24,25,26]:

- Compact size yielding low capital cost, modular construction, factory assembly, and low heat transfer area
- Higher thermal efficiency yielding lower thermal pollution
- Lower combustion temperature (1400°F-1800°F) yielding less fouling and corrosion
- Applicable to a wide range of low-grade fuels including char from synthetic fuels processes
- Adaptable to a high efficiency gas-steam turbine combined power generation cycle

The feasibility of the FBC for power generation depends in part on the following: development of efficient methods for regeneration and recycle of the dolomite/limestone materials used for sulfur absorption and removal; obtaining complete combustion through flyash recycle or an effective carbon burnup cell; development of a hot-gas particulate removal process to permit use of the combustion products in a combined-cycle gas turbine without excessive blade erosion.

The potential for reduced NO_x emissions with fluidized bed combustion is currently under investigation in several EPA funded projects. Preliminary tests with pilot scale units [24,25] indicated that emission levels well within the EPA standard of 0.7 lb $NO_2/10^6$Btu for a new coal-fired units can be achieved. At the operational temperatures of the fluidized bed, the rate of formation of thermal NO_x is very low and nearly all NO_x emitted results from conversion of fuel nitrogen. The fuel nitrogen content in the coals used in the pilot tests was not given, so these results cannot be generalized.

Several of the pilot scale units have been tested for the effects of operational variables on NO_x emissions. BCURA[26] has reported preliminary evidence that their pressurized fluidized bed yields lower emissions than their atmospheric unit. The bed temperature has little effect on NO_x emissions in the range from 1400°F-1800°F, but operation with excess air increases NO_x significantly. Argonne[25], and Esso[27], have suggested that operation with two-stage combustion may be effective for NO_x control in the firing of high nitrogen content coals. Esso suggests that two-stage combustion could have the additional advantage of increasing the efficiency of the sulfur removal process.

From a NO_x control standpoint fluidized bed combustion is regarded as medium risk concept because the economic feasibility of the basic process as well as NO_x control techniques has not been fully established relative to conventional boilers or low Btu gas combined-cycle units.

4.1.3.2 Gas Turbines and Reciprocating Engines

Control methods for new reciprocating I.C. engines and gas turbines include all of those techniques mentioned for retrofit (Section 4.1.1.2) plus those listed below.

I.C. Reciprocating	Turbines
Precombustion Chamber	Reduce Turbine Inlet Temperature
	Prevaporize Fuel
	Major Combustor Can Modification

There has been work in each of these areas which will be discussed in Section 5.4.2.2.

4.1.3.3 Other Combustion Sources

Other than the development of an optimum burner by Rocketdyne for home heating application, no new information was available on alternate processes for these other combustion sources for NO_x control.

4.1.4 Flue and Exhaust Gas Treatment

Esso reported in the 1969 report that of the various flue gas treatment schemes the aqueous scrubbing systems using alkaline solutions held the most promise. In a subsequent study by Chappel at Esso[28] under EPA sponsorship a number of aqueous solutions were investigated. The following conclusions are quoted from the Chappel report:

- The addition of NO_2 to flue gas to improve NO_x (mostly NO) absorption does not appear promising.

- Sulfite solutions and slurries are efficient NO_2-SO_2 absorbents. Soluble sulfites (Na_2SO_3) are better NO absorbers than insoluble slurries ($CaSO_3$).
- Calcium, magnesium and zinc hydroxide slurries are effective NO_2-SO_2 absorbers.
- Limestone ($CaCO_3$) is also a good NO_2-SO_2 absorbent.
- Sulfide solution are excellent NO_2-SO_2 absorbers but do generate a small amount of NO.
- Part of the absorbed SO_2 is oxidized to sulfate.
- Combined NO_x-SO_2 scrubbing seems feasible using any of several hydroxide or carbonate systems provided NO_x(NO) can be effectively oxidized by NO_2 upstream from the scrubbing unit.

The following recommendations were made for areas of future work.

"Pilot plant studies are needed to scale up the bench scale work developed in this study. The following critical factors need to be defined under process conditions:

- Rates of NO_2 absorbtion by different aqueous solutions under process conditions.
- Effect of scrubber design on absorbtion.
- Requirements for anti-oxidants must be assessed.
- The product distribution for the absorbed NO_x must be measured.
- Study of spent solution regeneration.
- Methods of optimizing NO_x to NO_2 oxidation by catalyst or ozone."

Although an exhaustive study was not made due to time limitations, to our knowledge there is still currently no established gas treating processes for NO_x control, especially for control of NO_x emissions from power plants. Reference is made to a new process developed by the Krebs Company in Section 4.2, Non-Combustion Sources, which is reported to be applicable to combustion sources in a combined SO_2-NO_x scrubbing scheme. However, to date it has not been applied to a full scale power plant. It is the general belief by most researchers that the primary control of NO_x emission will be through combustion modification and at some future time flue gas treatment schemes may be utilized to perform a secondary control, say to achieve 10-20 ppm of NO_x from a 60-100 ppm level.

4.2 NON-COMBUSTION SOURCES

Non-combustion sources include the following:

- Nitric Acid Plants
- Amonium Nitrate Manufacture
- Organic Oxidations
- Organic Nitrations
- Explosives
- Phosphate Rock Acidulation
- Metals treatment and etching

Nitric acid plants are the largest source of NO_x from these industries, although the other sources often produce intense local concentrations of NO_x, typically as a brown NO_2 plume. However, due to the relatively small contribution of these sources and the limited scope of this study, the control methods for these sources were not thoroughly investigated. It is our opinion that either scrubbing for NO_2 or process changes have been made on many of the sources to meet local ordinances for visible plumes. The reader is referred to References 29-32 for details on a number of processes. Reference 19 provides descriptive information on hundreds of U.S. patents since 1950 for flue gas treatment. The following areas are covered:

- Catalytic conversion of industrial stack gases
- Absorptive techniques for removing nitrogen oxides from gases
- Liquid scrub processes for removal of nitrogen oxides
- Combustion techniques for eliminating oxides in gas stream

In addition, a recent development by the Krebs Company is reported to achieve flyash, SO_2 scrubbing and NO_x scrubbing in a single apparatus. The system utilizes an aqueous solution of NaOH in conjunction with high efficiency, high surface area scrubbers, and a proprietary catalyst for NO, NO_2 conversion to N_2. It is reported applicable to any size source from metal etching plants to utility boiler and has achieved efficiencies of 99.5 percent for flyash, 93 percent SO_2 and 35-70 percent for NO_x. NO and NO_2 are converted to N_2 and N_2O in the process[33].

For existing nitric acid plants there are basically two methods that are being utilized as retrofit schemes, catalytic combustion and molecular sieve adsorption[22]:

Catalytic Combustion

This process involves the reaction of methane or hydrogen-rich fuel by NO_2 or NO and O_2 over a catalyst to produce CO_2, H_2O and N_2. The oxygen content of the tail gas is an important factor in determining the flow scheme of the catalytic combustion system, since the maximum oxygen removal per stage is approximately 3 percent. Therefore, above 3 percent oxygen either a two stage system is used or one stage with recycle. Most plants use a single stage unit and achieve only decoloration of the NO_2 to NO. By using additional stages, fuel, and catalyst all the NO_x can be abated but it is usually considered too costly. Several catalytic schemes are shown in Figure 4-2[34].

Molecular Sieve Adsorption

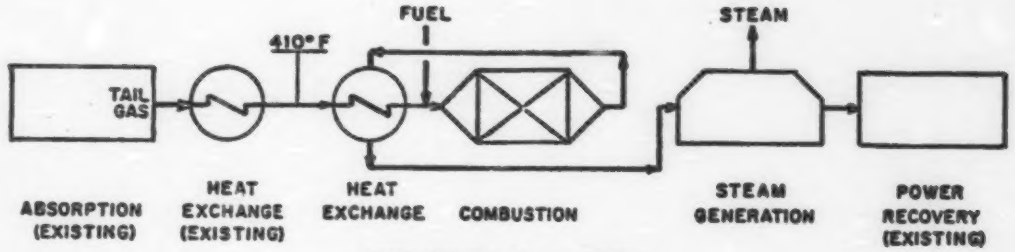
Union Carbide is currently working on a commercial molecular sieve adsorption scheme for NO_x removal from nitric acid plant tail gases. Concentration of less than 10 ppm are achievable. Figure 4-3 shows a typical schematic of the system. Two parallel streams are run to enable continuous operation while one stream is being regenerated. During the adsorption cycle the nitric oxide is catalytically converted to nitrogen dioxide, the tail gas water vapor is adsorbed and the total NO_x is adsorbed as NO_2. During regeneration the nitrogen dioxide is recycled to the nitric acid absorption equipment. Proponents for the molecular sieve scheme claim compliance with projected EPA regulations. It may also be more attractive due to less fuel consumption.

Table 4-1 presents some typical costs for the two schemes based on a TVA nitric acid plant[34]. Union Carbide reports costs for their scheme of approximately \$1.50/ton of HNO_3 for a 300 ton/day plant. These costs are changing rapidly due to increased cost of stainless steel.

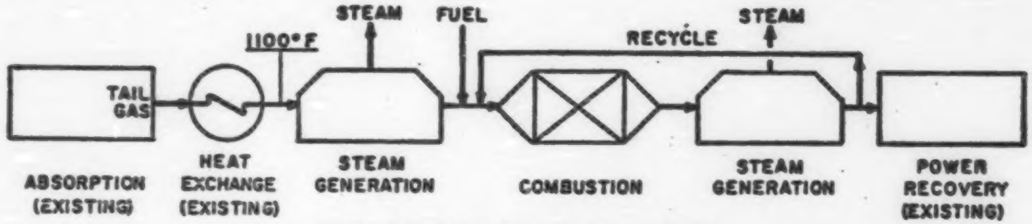
4.3 SUMMARY

A summary evaluation of NO_x control options is given in Table 4-2. For the control of existing combustion sources, combustion modification has remained, since the Esso study, as the quickest, most reliable and cost effective technique. For non-combustion sources, flue gas treatment is preferred.

For long term NO_x control, the incorporation of combustion modifications into new unit design appears to be the single most effective strategy for stationary sources. Large electric power generation units will undoubtedly benefit from developments in the use of synthetic fuels and fluidized bed combustion in conjunction with combined power cycles. Even these advanced concepts, however, may require combustion modification for NO_x control. The bulk of stationary sources — area sources such as pipeline I.C. engines, industrial


FUEL
STEAM
410°F
TAIL GAS
ABSORPTION (EXISTING)
HEAT EXCHANGE (EXISTING)
HEAT EXCHANGE
COMBUSTION
STEAM GENERATION
POWER RECOVERY (EXISTING)

ONE-STAGE ($O_2 < 3\%$)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RECYCLE
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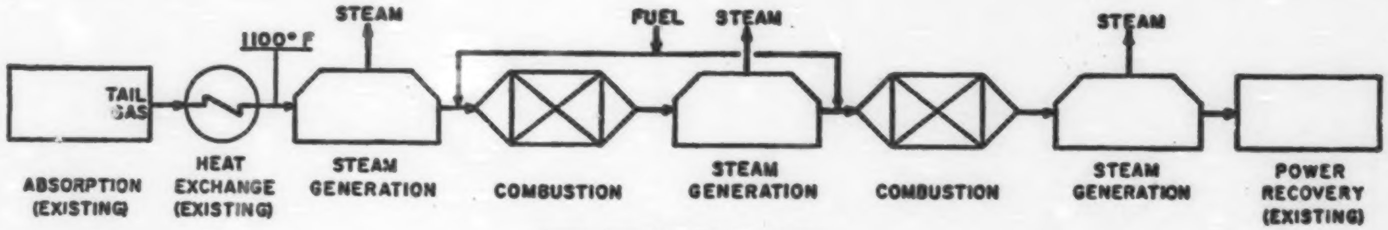
TAIL GAS
ABSORPTION (EXISTING)
HEAT EXCHANGE (EXISTING)
STEAM GENERATION
COMBUSTION
STEAM GENERATION
POWER RECOVERY (EXISTING)

ONE-STAGE RECYCLE ($O_2 > 3\%$)


STEAM
FUEL
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STEAM
1100°F
TAIL GAS
ABSORPTION (EXISTING)
HEAT EXCHANGE (EXISTING)
STEAM GENERATION
COMBUSTION
STEAM GENERATION
COMBUSTION
STEAM GENERATION
POWER RECOVERY (EXISTING)

TWO-STAGE ($O_2 > 3\%$)

Figure 4-2. Catalytic Combustion Systems

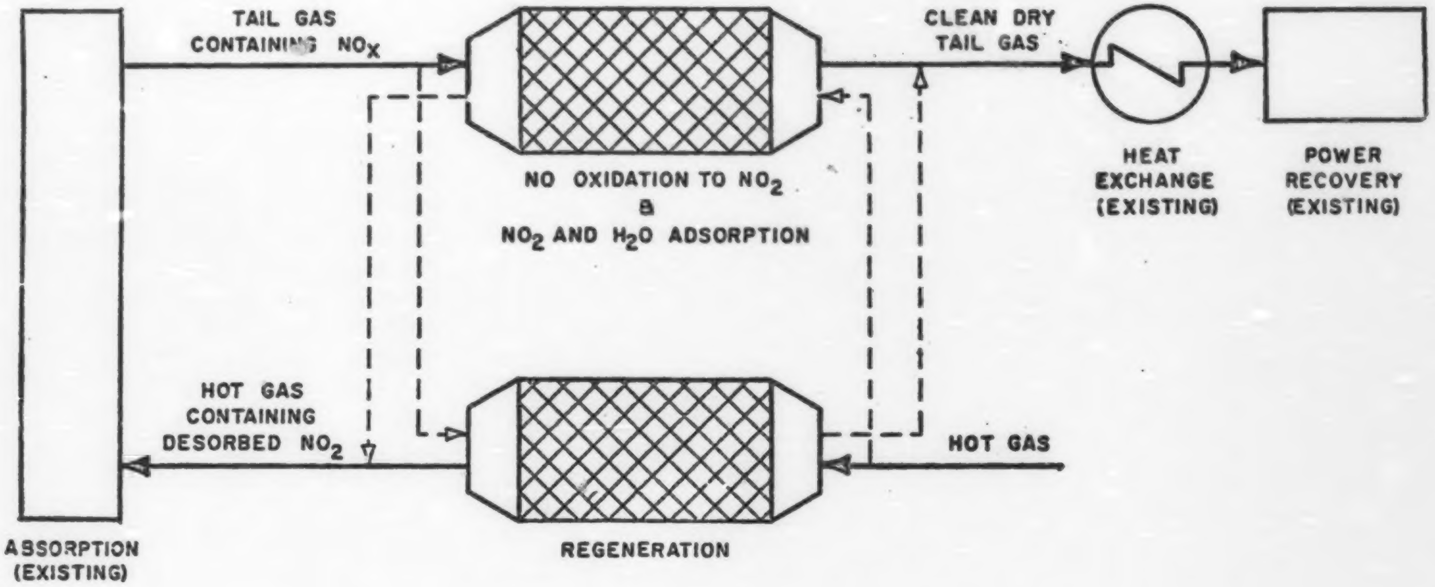
TAIL GAS
CONTAINING NO_X
CLEAN DRY
TAIL GAS
HEAT
EXCHANGE
(EXISTING)
POWER
RECOVERY
(EXISTING)
NO OXIDATION TO NO_2
&
NO_2 AND H_2O ADSORPTION
HOT GAS
CONTAINING
DESORBED NO_2
HOT GAS
ABSORPTION
(EXISTING)
REGENERATION

Figure 4-3. Molecular Sieve System

TABLE 4-1
COST FOR NO_x ABATEMENT FACILITIES FOR THE NO. 3 NITRIC ACID UNIT AT TVA

	Catalytic Combustion Using Natural Gas[a]	Molecular Sieve Adsorption
Approximate Investment Cost, $	290,000	675,000
Depreciation Cost (10 yr.), $/Ton HNO_3	0.53	1.24
Approximate Operating Costs, $/Ton HNO_3		
Natural Gas	1.48	0.07
Electricity	--	0.11
Steam	--	0.06
Cooling Water	--	0.04
Reduction Catalyst	0.10	--
Molecular Sieve	--	0.26
Operation, Overhead, and Maintenance	1.00	1.00
Byproduct Credits		
Steam	-2.04	--
Nitric Acid	--	-0.25
Total Operating Cost	0.54	1.29
Total Estimated Cost for NO_x Abatement, $/Ton HNO_3[b]	1.07	2.53

[a] Data are for two-stage or partial recycle-type combustion process.

[b] Total cost for NO_x abatement is the sum of the project cost with 10 year depreciation and the operating costs.

TABLE 4-2

SUMMARY OF NO_x CONTROL OPTIONS

Option	Approach to NO_x Control	Effectiveness		Overall Evaluation
		Short Term	Long Term	
Combustion Modification	Suppress formation of thermal, fuel No_x by modifying operating conditions	Present applicability; preferred option	Incorporate into optimized low NO_x design	Priority option for short and long term control of boilers, furnaces, turbines
Synthetic Fuels	Reduced flame temperatures from low Btu gas yields reduced NO_x	Negligible application	Promising low risk concept for post-1980	Attractive long term option; May require combustion modifications
Fuel Additives	Reduce or decompose NO to N_2 by catalytic addition	Not effective	High risk development	Not promising
Fluidized Bed Boiler	Low bed temperature suppresses thermal NO_x	N.A.	Medium risk concept for post-1980	Promising long term option; May require combustion modifications
Flue or Exhaust Gas Treatment	Reduce NO to N_2 by catalytic treatment, or absorption of NO or NO_2.	Preferred for nitric acid plants	Possible for control of I.C. Engines	Potential long-term option for backup of combustion modification for boilers, furnaces. Possible long-term option for I.C. Engines. Priority option for nitric acid plants.

combustion, and domestic heaters — will increasingly utilize combustion modifications for NO_x control in new equipment design.

Combustion modifications are clearly the priority item in program planing for NO_x control of stationary sources. In order to indicate requirements for effective utilization of combustion modifications, the next section characterizes the current status of combustion control in the light of developments since the Esso study.

SECTION 5

COMBUSTION MODIFICATION TECHNOLOGY FOR NO_X CONTROL

The Esso systems survey detailed an R&D plan for which the priority category was the development of combustion modification technology. The recommendations focused on pilot scale and full scale testing of combustion modification techniques with support from fundamental studies on the combustion process and NO_X formation mechanism. Subsequently the EPA has organized a Combustion Control Program to develop and coordinate R&D for NO_X control; the Esso recommendations have largely been treated as part of the operation of this program. In this chapter, a review is made of the achievements, current status, and prospects for combustion control technology. The objective is to specify areas of development required for program planning. Specific R&D recommendations follow in Section 6.

The basic objective of combustion modification technology is to discover and apply techniques which alleviate NO_X formation. A review of known factors involved in the problem of thermal and fuel NO_X formation is given in Section 5.1. A combustion control program directed at confronting the problem is described in Section 5.2. The status and limitations are given in 5.3 for fundamental studies, and in 5.4 for pilot and full scale testing. Representative cost data for combustion modifications are given at the end of 5.4.

5.1 COMBUSTION GENERATED NO_X

Oxides of nitrogen formed in combustion processes are usually due either to thermal fixation of atmospheric nitrogen in the combustion air, leading to thermal NO_X, or to the conversion of chemically bound nitrogen in the fuel, leading to fuel NO_X. Any coupling between these two mechanisms is thought to be minor so that total NO_X emissions are essentially the sum of thermal and fuel NO_X. For natural gas and light distillate oil firing, all NO_X is formed via atmospheric fixation. With residual, or crude oil, and coal, the contribution from fuel bound nitrogen can be significant and, under certain operating conditions, predominant.

A third potential mechanism of NO_x formation arises in processes such as glass manufacturing, where the raw materials in contact with the combustion products contain nitrogen compounds. Little is known about the extent of conversion to NO_x of the nitrogen compounds, or of the effects of combustion modifications on this mechanism.

5.1.1 Thermal NO_x

The detailed chemical mechanism by which molecular nitrogen in the combustion air is converted to nitric oxide is not fully understood. In practical combustion equipment, particularly for liquid or solid fuels, the kinetics of the N_2-O_2 system are coupled to the kinetics of hydrocarbon oxidation and both are influenced, if not dominated, by effects of turbulent mixing in the flame zone. It is, however, generally accepted that the most significant reactions in the formation of thermal NO_x are those of the Zeldovich chain mechanism:

$$O_2 + M \rightleftarrows O + O + M \ , \tag{5-1}$$

$$N_2 + O \leftrightharpoons NO + N \ , \tag{5-2}$$

$$O_2 + N \leftrightharpoons NO + O \ , \tag{5-3}$$

for which reaction (5-2) is the rate controlling mechanism. "M" is a third body, normally taken as N_2. Additional mechanisms are reviewed in Section 5.3.2.

Due principally to the high energy required to break the N_2 bond in (5-2), the activation energy for NO formation via the Zeldovich mechanism is considerably larger than for typical rate-controlling reactions in hydrocarbon oxidation. This entails that NO formation is initiated well after initiation of fuel combustion and is extremely temperature sensitive with virtually all NO being formed in the high temperature regions of the flame. For the time scales involved in the flow through commercial combustors, the high temperature dependence of the NO system means that total NO emissions are far below equilibrium levels. NO formation is thus kinetically controlled with the emission level dependent on time of exposure to the high temperature.

Figure 5-1 illustrates the temperature and time dependencies of NO formation for an idealized one-dimensional premixed system of methane and air[35]. The results at 0.01 sec for three values of stoichiometric ratio (S.R.)

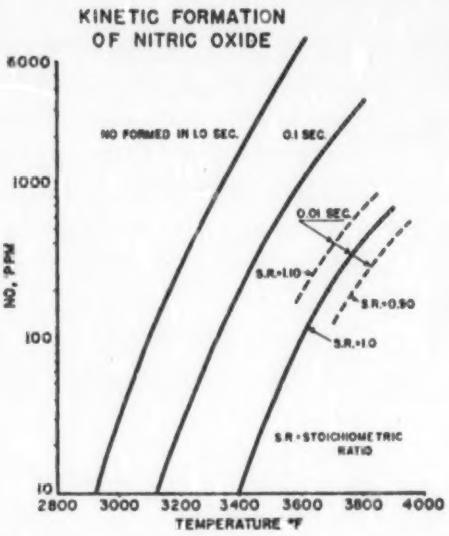

Figure 5-1. Kinetic Formation Of Nitric Oxide

show, as expected, that NO formation is suppressed by reduced availability of oxygen. In a practical combustor, departure from S.R. = 1 would result in reduced temperatures which would further suppress NO formation. It is precisely these factors of high sensitivity to temperature, concentration level, and time of exposure which makes the formation of thermal NO_x susceptible to the combustion modification strategy.

Ideally, the formation of thermal NO_x could be reduced by four tactics:

- Reduce nitrogen level at peak temperature
- Reduce oxygen level at peak temperature
- Reduce peak temperature
- Reduce time of exposure at peak temperature.

Esso showed that reduced nitrogen level is impractical so the strategy has focused on the fundamental parameters of oxygen level, peak temperature, and time of exposure. These parameters are in turn dependent on secondary combustion variables such as combustion intensity and internal mixing in the flame zone - effects which are ultimately determined by primary equipment and fuel parameters over which the combustion engineer has some control. A hierarchy of effects leading to thermal NO_x formation is depicted on Table 5-1.

TABLE 5-1

FACTORS CONTROLLING THE FORMATION OF THERMAL NO_x

Primary Equipment and Fuel Parameters	Secondary Combustion Parameters	Fundamental Parameters	
Inlet temperature, velocity Firebox design Fuel composition Injection pattern of fuel & air Size of droplets or particles Burner swirl External mass addition	Combustion intensity Heat removal rate Mixing of combustion products into flame Local fuel-air ratio Turbulent distortion of flame zone	Oxygen level Peak temp. Exposure time at peak temp.	Thermal NO_x

Although causal relationships between the four categories shown in Table 5-1 are not firmly established, combustion modification technology is, nevertheless, confronted with the task of reducing thermal NO_x through modification of equipment and fuel parameters. This task has been approached with efforts ranging from the short-term testing of equipment modifications on commercial units, in order to determine the effects on NO_x emission, to long-term fundamental studies and pilot testing directed at achieving a basic understanding of NO_x formation. The Combustion Control Program devised to coordinate these efforts is described in Section 5.2.

5.1.2 Fuel NO_x

Since the Esso study, there has been increasing recognition of the role of fuel bound nitrogen in NO_x emissions. With increasing utilization of coal, particularly high nitrogen coal, fuel NO_x control has become a priority item in combustion modification technology.

When a droplet or particle of fuel is injected through a flame, some, and possibly most, of the fuel nitrogen compounds go towards the formation of intermediate nitrogen compounds, such as HCN or NH_3, in the gas phase volatilization products. The remainder of the nitrogen stays in the char or soot and may be converted during carbon burnout. A number of fuel bound nitrogen compounds have been identified,[36,37] but the degree of conversion to NO_x does not appear to be strongly dependent on the type of compound[36,37,38]. The percent of fuel nitrogen which gets converted to NO is strongly dependent on the combustion conditions, but generally decreases as the percentage of nitrogen in the fuel is increased.

In contrast to thermal NO_x, fuel nitrogen conversion is generally regarded as being relatively insensitive to temperature. This may be due to the relatively low energies involved in the formation of the intermediate compounds from the fuel bound compounds. The most critical factor in fuel NO_x conversion appears to be the local conditions in which volatilization and formation of intermediate compounds occurs. In a reducing atmosphere, it is suspected that the intermediate compounds go to form N_2 or other compounds with little subsequent conversion to NO. In an oxidizing atmosphere, conversion of the intermediates to NO is thermodynamically favored over conversion to N_2. Although basic understanding of these phenomena is only in the preliminary stage, a promising strategy for fuel NO_x reduction appears to be modification of the burner or combustion conditions to allow volatilization to occur prior to massive entrainment of oxygen in the flame zone.

The fate of fuel-bound nitrogen which does not go to NO is uncertain. There are indications that other pollutants, such as HCN, may result when NO formation is suppressed[40,41]. This possibility may, indeed, constitute a limitation to fuel NO_x reduction strategies and requires investigation.

5.2 SYSTEMS PROGRAM FOR COMBUSTION MODIFICATIONS

A combustion modification program to attack the problem of NO_X formation must satisfy both the short term requirement for immediate application of combustion modification to existing units, and the long term requirement for generation of optimized low NO_X design concepts for new units. These requirements must be met in the face of difficulties such as: uncertainties in NO_X formation mechanisms; lack of causal relation between equipment parameters and the formative mechanisms; lack of reliable laws for scaling pilot scale data to full scale units; prohibitive time and cost of exhaustive full scale testing. In view of these factors, the EPA, based partly on the Esso recommendations, has formulated an R&D program involving fundamental studies, pilot scale testing and full scale testing. The program is organized into four component areas:

Fundamental Studies

Fuels R&D

Process R&D

Field Testing and Survey

Fundamental Studies involves bench scale experimentation as well as analytical modeling to attain a better understanding of the physical and chemical phenomena in combustion and NO_X formation. This understanding provides a rational basis on which to evaluate data and trends from pilot and full scale testing. Fundamental studies also contribute to the generation of basic ideas for potential combustion modifications and aid in the application and optimization of combined modifications. Finally, the basic understanding of the combustion process can be useful in combating operational difficulties resulting from modifications and hence extends the effectiveness of the modification strategy.

Fuels R&D involves the generalized testing of combustion modifications on non-commercial, versatile, laboratory test equipment. Its presence in the R&D program is necessitated by the prohibitive cost of diversified testing of full scale units. Fuels R&D functions as a screening of candidate modification techniques in order to evaluate and optimize generalized procedures and detect limitations or operational difficulties. Fuels R&D occupies a middle position between the basic chemical and physical focus of fundamental studies and the hardware focus of process R&D and field testing.

Process R&D involves demonstration testing of modification techniques on units with hardware which is characteristic of commercial systems. It functions in the development of operational guidelines for implementation of combustion modifications in commercial equipment, and serves to detect and solve operational difficulties.

Field Testing and Survey involves the continuing definition and application of state-of-the-art technology for full-scale commercial equipment. The results from this effort go directly to industrial application for controlled emissions.

The functioning and interrelationships of the component areas of the modification program are depicted schematically in Figure 5-2. Field testing and survey is the central short-term focus of the program and operates parallel to the supporting areas of fundamental studies, fuels R&D, and process R&D. Field testing benefits from information generated in the supporting areas and provides input to these areas in the form of test data and information on operational difficulties, limitations, and cost effectiveness. Thus, the directional requirements of R&D in the four component areas are continually redefined in view of results from the other areas. This yields a flexible base on which to respond to changing requirements in short and long term objectives.

5.3 APPLICATION OF FUNDAMENTAL STUDIES

Since the Esso study, the most significant developments impacting the fundamental studies area are the increasing importance of the fuel NO_x problem, and the increasing use of computerized analysis of basic physical and chemical phenomena. These developments have broadened the scope of fundamental studies and have somewhat reoriented the strategy used for support of short- and long-term requirements. This section characterizes the application to combustion modification technology of fundamental studies in order to give directional requirements for R&D planning.

Fundamental studies involves experimentation and analysis aimed at understanding the NO_x formation process. The incorporation of these elements into a simulation of the actual combustion process is termed modeling. The scope of modeling extends from simple scaling relations for extrapolating pilot scale tests, to a mathematical simulation of complex phenomena such as hydrocarbon oxidation or fuel nitrogen conversion. Models are the long-term end product of fundamental studies in the sense that they represent the best available rationalization derived from analysis and experimentation.

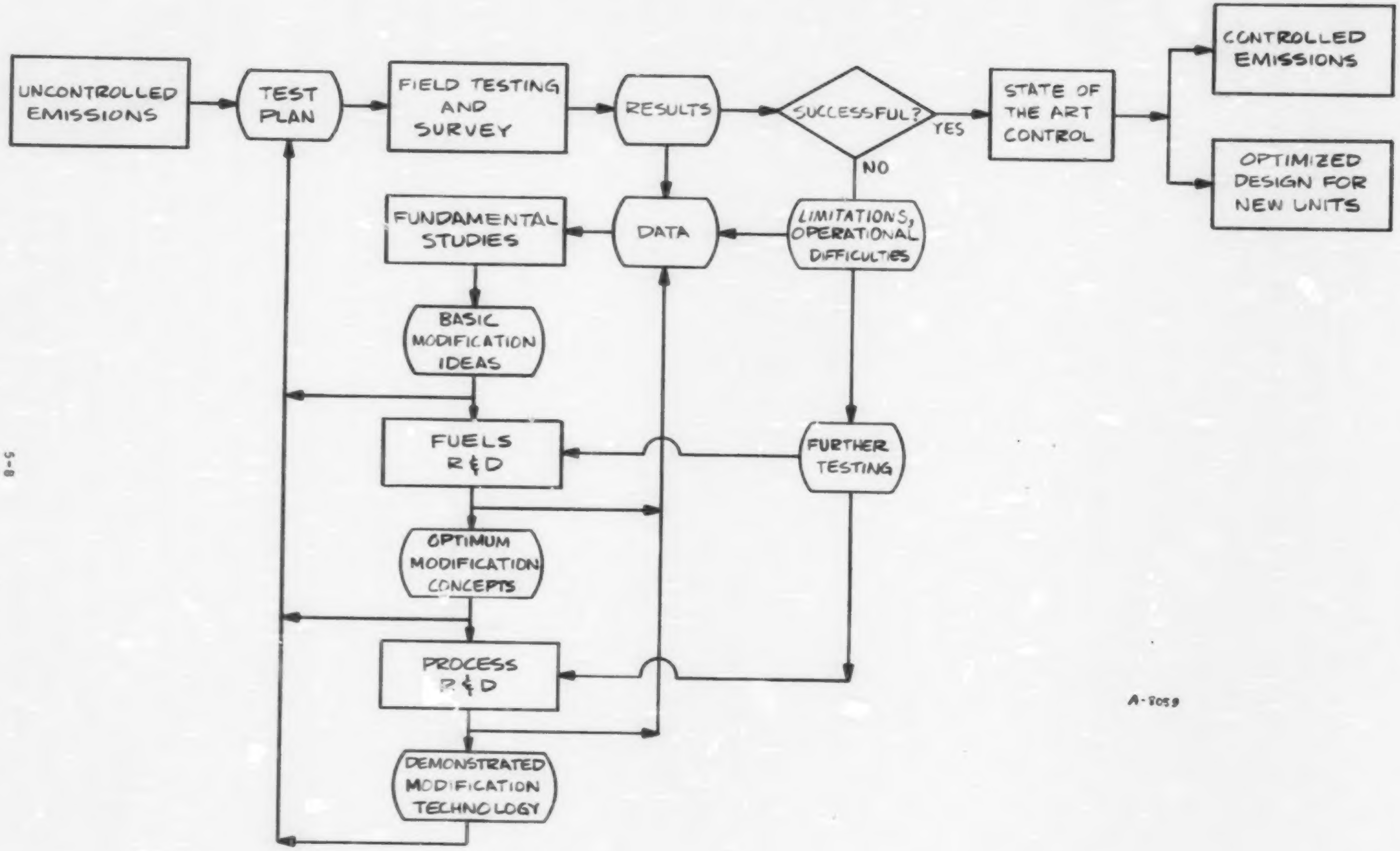
UNCONTROLLED EMISSIONS
TEST PLAN
FIELD TESTING AND SURVEY
RESULTS
SUCCESSFUL?
YES
NO
STATE OF THE ART CONTROL
CONTROLLED EMISSIONS
OPTIMIZED DESIGN FOR NEW UNITS
DATA
LIMITATIONS, OPERATIONAL DIFFICULTIES
FUNDAMENTAL STUDIES
BASIC MODIFICATION IDEAS
FUELS R&D
FURTHER TESTING
OPTIMUM MODIFICATION CONCEPTS
PROCESS R&D
DEMONSTRATED MODIFICATION TECHNOLOGY

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Figure 5-2. The Combustion Control Program for NO_x Control

5.3.1 Strategies for Model Development

On a short-term basis, the principle functions of modeling are to generate basic ideas for combustion modifications, guide in the experimental planning of pilot tests, and to provide a rational basis for data evaluation. The long-term function includes definition of the minimum attainable emission levels, optimization of combustion modifications, and ultimately, predictions for use in low NO_x designs. Since the exploratory modeling study of Esso[1], there have been extensive experimental and analytical efforts directed at fulfilling both the short- and long-term function of modeling. As would be expected for an inherently long term program, much work remains for the full utilization of the potential of fundamental studies. The experience to date, however, does offer valuable guidelines for facilitating further development. From a systems perspective, strategies for coordinating model development include:

- Simultaneous focus on experimentation and analysis,
- Integration of short and long term requirements,
- Simultaneous development of auxiliary relations,
- Consistent level of sophistication in models,
- Use of complex models to calibrate simple models,
- Coupling of combustion and NO_x modeling.

Some uses and implications of these strategies are summarized on Table 5-2.

At the time of the Esso study, the emphasis in fundamental studies was on physical modeling which, typically, avoids consideration of basic phenomena, such as turbulent mixing, thermal radiation, etc., through use of restrictive physical assumptions. As delineated in Table 5-2, the recent emphasis in fundamental studies is on formulation of auxiliary models for each of the key phenomena contributing to NO_x formulation. These are:

- Fluid flow phenomena
- Hydrocarbon combustion
- Thermal NO_x
- Fuel NO_x
- Turbulent viscosity
- Turbulent mixing effects on kinetics
- Thermal radiation
- Droplet or particulate flow and combustion.

TABLE 5-2

TYPICAL STRATEGIES FOR MODEL DEVELOPMENT

Strategy	Description	Examples	Possible Future Applications
Coordinated Experimentation and Analysis	Computerized analysis has refocused priority on experimentation. Data inititiates, refines and verifies models, so experimental planning requires coordination with modeling needs (Fig. 5-3).	Modeling of swirl effects in gas turbine combustors (UARL); identification of key species in hydrocarbon flames (Esso, Ultrasystems)	Turbulent mixing effects on combustion and NO_x formation in jet diffusion flames. Modeling of pulverized coal flames
Integration of Short and Long Term Requirements	Short term requirements are a natural fallout of long term program (Fig. 5-3)	UARL flame modeling yields useful flow pattern predictions	Fuel nitrogen modeling to generate new ideas for combustion modifications
Simultaneous Development of Auxiliary Models	Focus modeling at a basic level to identify effects on NO_x formation	Current programs on H/C kinetics, NO_x kinetics, radiation, etc.	Expansion of fuel nitrogen modeling efforts; modeling of turbulent mixing effects on H/C and NO kinetics
Use of Complex Models to Calibrate Simple Models	Develop approximate techniques from results of single or coupled auxiliary models	Global H/C kinetics from detailed kinetic calculations; simple flow models from recirculating flow code; correlations for luminous radiation	Simple eddy mixing models for effects of turbulent mixing on reactions
Coupling of NO_x Modeling and Combustion Modeling	H/C and NO_x kinetics are coupled in many combustors; turbulent mixing dominates reaction rates in practical systems, so NO_x predictions must be coupled to combustion predictions	Modeling of flame-zone NO_x formation (Ultrasystems, Esso)	Coupling of combustion aerodynamics and NO_x modeling (UARL)

In the short term, these auxiliary models are used to generate ideas for combustion modification concepts, and to develop understanding of specific aspects of the NO_x formation process. In the long term, two more auxiliary models can be coupled to relate fuel or equipment parameters to NO_x formation.

The evolutionary development of NO_x modeling through the above approach is depicted on Figure 5-3. The current emphasis is on formulation of first generation models for the key items such as fuel NO_x and hydrocarbon combustion. The ultimate utility of this approach is contingent upon development of the auxiliary models to a consistent level of sophistication. The status and prospects of auxiliary model development is reviewed in the following section.

5.3.2 Status and Prospects of Modeling

The priority area in fundamental studies is the investigation of the basic phenomena such as NO_x kinetics, fuel NO_x, and thermal radiation involved in NO_x formation. This information is necessary to relate NO_x formation to actual combustion modifications and, effectively, fill in the gaps in Table 5-1.

Table 5-3 gives a summary of status and prospects of auxiliary models related to NO_x formation. The final column contains a speculation on the probable optimum level of development for use in data correlations and predictions. The status of these individual areas is reviewed below. This is not intended to be an exhaustive treatment of current research, but only an indication of requirements for R&D planning.

Fluid Flow Solutions

This is the most advanced area of combustion modeling primarily because the problems in code development are mathematical rather than physical and thus do not require experimentation. Early fluid flow solutions, used for example, by Esso[1,38] and IFRF[42], employed plug flow or well stirred reactor concepts. It was found necessary to place several of these elements in a series or parallel arrangement in order to approach a realistic simulation of practical flows. This added complexity required ever increasing empirical specification to link the various elements. Esso[38] and IFRF[42] reported unsatisfactory results with this approach. The use of well-stirred or plug-flow elements to model gas turbine combustors has been more promising[43].

The numerical flow solution is a natural extension of the use of series and parallel well-stirred reactors or plug flow elements. With the numerical appraoch, the flow field is divided into a large number of computation cells,

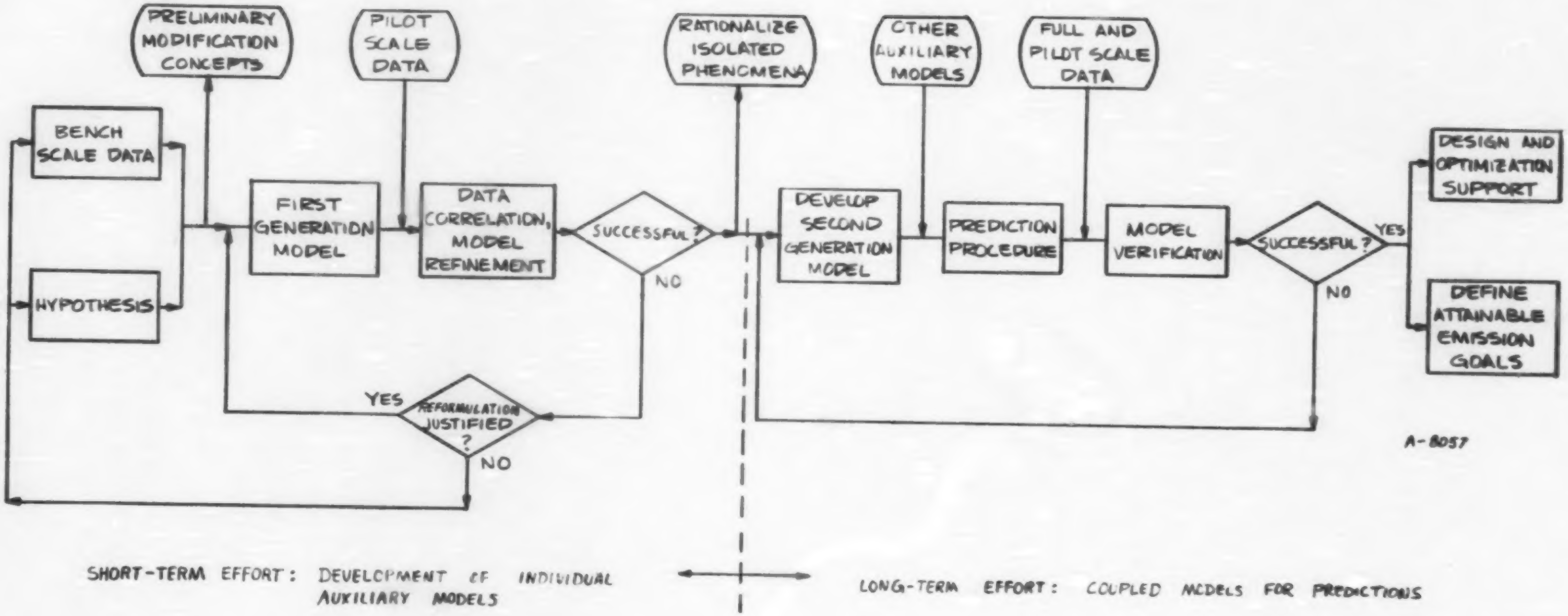

PRELIMINARY MODIFICATION CONCEPTS
PILOT SCALE DATA
RATIONALIZE ISOLATED PHENOMENA
OTHER AUXILIARY MODELS
FULL AND PILOT SCALE DATA
BENCH SCALE DATA
HYPOTHESIS
FIRST GENERATION MODEL
DATA CORRELATION, MODEL REFINEMENT
SUCCESSFUL ?
NO
REFORMULATION JUSTIFIED ?
YES
NO
DEVELOP SECOND GENERATION MODEL
PREDICTION PROCEDURE
MODEL VERIFICATION
SUCCESSFUL ?
YES
NO
DESIGN AND OPTIMIZATION SUPPORT
DEFINE ATTAINABLE EMISSION GOALS
A-8057
SHORT-TERM EFFORT: DEVELOPMENT OF INDIVIDUAL AUXILIARY MODELS
LONG-TERM EFFORT: COUPLED MODELS FOR PREDICTIONS

Figure 5-3. Typical Sequence in Model Development

TABLE 5-3

STATUS OF MODEL DEVELOPMENT

Area	Current Capability	Advanced Stage of Development	Preliminary Stage of Development	Estimated Best Use in Practical Predictions
Fluid Flow Solution	Plug flow; stirred reactor; 2-D Boundary Layer	2-D Recirculating flow with swirl; 3-D Boundary Layer	3-D Recirculating flow	2-D Recirculating flow
Gas-phase Hydrocarbon Combustion	Equilibrium diffusion flames; premixed flames with global kinetics	Multi-step reaction for methane	Multi-step reaction for complex hydrocarbons	Multi-step reactions for complex hydrocarbons
Thermal NO Kinetics	Zeldovich Mechanism	Extended Zeldovich Super Equilibrium prompt NO	Coupled NO and hydrocarbon kinetics	Coupled NO and hydrocarbon kinetics
Turbulent Viscosity	Algebraic mixing length	One- and two-equation models for boundary layers	Two equation models for recirculating flow; multi-equation Reynolds stress models	Two equation models for swirling flows
Mixing Effects on Kinetics	Neglect mixing; time-averaged temperature, concentration used in kinetic calculation	Simple eddy mixing models for flame zone	Statistical treatment of temperature, concentration fluctuation effects on reaction rates	Eddy mixing models for rates of combustion and NO formation
Thermal Radiation	Gas phase radiation simple geometries	Simple correlations for luminous radiation; gas radiation in complex geometries	Models for luminous radiation	Combined gaseous, luminous radiation models for complex geometries
Fuel Nitrogen	--	--	Modeling of volatilization and heterogeneous fuel NO_x conversion, single particles	Fuel NO_x models for pulverized coal flames
2ϕ Droplet, Particulate Combustion	--	Non-reacting 2-D droplet/particulate flow; single droplet combustion	Models for multi-droplet/particulate flames	Droplet/particulate density and size distribution for use in fuel NO_x and luminous radiation models

each of which can be considered as a well-stirred reactor. The advantage of this approach is that the mass and energy transport between cells satisfies the governing mass, momentum, and energy balances.

Two dimensional boundary layer codes have achieved a near-optimum state of development and are in routine use for a variety of flows including axi-symmetric flames with small swirl[44]. These codes can be exploited in the EPA fundamental studies through use in predicting jet flames, with or without swirl, to test models for turbulent viscosity, radiation, two phase flow, and effects of turbulent mixing on flame propagation.

Two-dimensional recirculating flow codes in present use are mostly based on the Imperial College vorticity-stream function (ψ-ω) code[45]. Differing versions of this type of code are possessed by Aerotherm, Battelle, KVB Engineering, UARL, and Ultrasystems. UARL is currently doing an important study, including experimentation, to model swirling reacting flows in combustors. Further development of the basic (ψ-ω) code will be mainly in the areas of numerical stability and computing time reduction. There has also recently been developed a pressure-velocity (p-v) code for 2-D recirculating flows[46]. This code has considerable advantage over the (ψ-ω) code, particularly in boundary condition specification and computational efficiency, because the solution is made with the physical variables. The (p-v) method is also readily extended to 3-D flow[47], whereas the (ψ-ω) method is not. The 3-D codes are useful for obtaining crude predictions of furnace flow patterns, but accurate flame zone predictions are precluded by uncertainties in turbulence and other parameters.

Hydrocarbon Oxidation

For prediction of combustion in hydrocarbon premixed flames, it is necessary to reduce the numerous elementary reactions involved to a manageable set of controlling reactions which simulate the actual process. EPA is currently funding three related studies which will lead to a better definition of the important reactions and the kinetic constants for both hydrocarbon oxidation and NO formation. Esso R&E is measuring concentrations in premixed flames in order to identify important species and mechanisms in the flame zone. Ultrasystems is doing a corresponding modeling study to reduce the detailed methane kinetics to a small number of controlling reactions. Part of this effort involves correlation of the Esso data. SRI is conducting a supporting study to develop prediction methods for the kinetic constants for elementary reactions. These results are useful in determining rate controlling or global reactions. These three studies are contributing to the basic understanding of combustion and NO_x formation which is required for modeling of complex hydrocarbon combustion and the coupling with NO_x formation.

For many practical combustors, the flames are of diffusion type for which the mixing rate of fuel and oxidizer controls the heat release rate. Nitric oxide emission patterns for diffusion flames can be quite different, especially in the dependancy of fuel-air ratio, than for premixed flames[48,49]. This factor, coupled with the importance of diffusion flame phenomena in fuel nitrogen conversion, gives diffustion flame studies a level of importance comparable to premixed flame studies.

Thermal NO Mechanisms

There has been extensive testing of the degree of coupling between NO kinetics and combustion kinetics under various combustion conditions. The precise conditions under which alternate mechanisms apply is not fully understood. Proposed mechanisms are reviewed below in order of increasing complexity.

i) Zeldovich with Equilibrium Concentration of Oxygen Atoms, [0]: This is the classical mechanism by which NO formation proceeds according to:

$$O_2 + M \rightleftharpoons 2O + M \ , \ [O] = K_{eq} \, [O_2]^{1/2} \ , \tag{5-4}$$

$$N_2 + O \rightleftharpoons NO + N \ , \tag{5-5}$$

$$O_2 + N \rightleftharpoons NO + O \ , \tag{5-6}$$

and for which reaction (5-4) is in equilibrium. This mechanism has been applied extensively especially for I.C. engines, and the best success has been obtained in the post-flame region of fuel-lean high pressure flames.

ii) Zeldovich with Non-equilibrium [0]: Fenimore[50] noted that reactions (5-4) - (5-6), with equilibrium [0], failed to predict NO formations near the flame zone. The "prompt NO" phenomenon has since been observed by several other investigators[51 - 54]. Bowman[55] and others[51 - 53] have suggested that prompt NO is due to non-equilibrium [0] concentrations in the near-flame region. Use of (5-5) and (5-6) with super-equilibrium levels of [0] has given reasonable correlations of prompt NO in many cases. It has yet to be determined when this approach is sufficient to explain prompt NO, and, also, what constitute satisfactory approximations for the non-equilibrium concentration of [0].

iii) Extended Zeldovich: For fuel rich flames, the concentration of the [OH] radical in the near flame region is sufficiently high to give importance to an additional reaction:

$$N + OH \rightleftharpoons NO + H. \qquad (5\text{-}7)$$

This reaction has been used both with the equilibrium oxygen atom assumption as well as the super-equilibrium [O] correction. The contribution of (5-7) to NO formation is far less than from the Zeldovich reactions, but its inclusion for fuel-rich flames seems necessary.

iv) Fully Coupled NO_X/HC Kinetics: Fenimore[50] and Iverach[54] have proposed that [N] atoms participating in the Zeldovich reactions may, in part, be due to reactions with flame radicals such as:

$$CH + N_2 \rightarrow HCN + N \ ,$$

$$C_2 + N_2 \rightleftharpoons 2CN \ ,$$

$$C + N_2 \rightleftharpoons CN + N \ .$$

The validity or conditions of applicability of these reactions has not been fully established, but it is generally agreed that in fuel rich, low pressure flames, NO_X kinetics are coupled to the combustion kinetics. The Esso and Ultrasystems studies mentioned above are looking at the coupling mechanisms which exist in the flame zone. An additional coupling, that between NO_X and sulphur dioxide, is being studied by the University of Arizona.

Turbulent Viscosity

Flow predictions in swirling flame zone requires modeling of the turbulent viscosity. Following the development of flow prediction codes, it became possible to test and refine turbulence models in complex flow situations. Routine use is now being made of algebraic mixing length models[56,57], and one- or two-equation models[58,59,60] in which differential equations are solved for turbulence parameters such as turbulent kinetic energy or the length scale. For recirculating flows, these models are normally used with the assumption that the turbulence is locally isotropic. For high rates of swirl, it has been shown[61] that the viscosity in the swirl direction differs radically from the main flow direction, and the locally isotropic assumption is not adequate for predictions. Testing of viscosity models for low and high rates of swirl would be useful.

Multi-equation Reynolds stress models are being developed by the Los Alamos group and the Imperial College group. These are costly to compute and this level of complexity is usually not required. Their chief contribution to combustion modeling will probably be to calibrate simple one- or two-equation models.

Turbulent Mixing Effects on Reaction

The distortion of the flame zone due to turbulence has been identified as a high priority study area by several investigators. In turbulent flames, the local temperature and concentration exhibit large fluctuations due to the mixing of turbulent eddies. The rate of combustion and NO_X formation are highly non-linear functions of temperatures and concentration; use of time average values yields erroneous flame propagation and NO_X formation predictions.

Predictions of the effects of turbulent mixing is still in a preliminary stage of development. Several eddy-mixing models have been used, either for combustion reaction rates[62,63], or for NO_X formation[64], with some success. MIT[65] has proposed a statistical approach for the specification of the composition of eddies for use in prediction of combustion and NO_X formation.

Thermal Radiation

Thermal radiation is of particular importance in NO_X formation from utility and industrial boilers, but less so for I.C. engines and domestic heaters. Modeling activities are focused both on methods for calculating radiative transport and on emissivity predictions for gases and luminous flames.

For furnace heat transfer predictions, the zone method[66] and the two or four flux models[67,69] have both been applied, with the flux models usually preferred for use in numerical flow codes. Radiative properties for the gas phase combustion products are known to an adequate degree for most cases[68]; for luminous flames, the radiation is dominated by scattering and particulate density, and the properties are less developed[69]. The IFRF has been extensively involved in measuring flame radiation and modeling luminous emissivities. Battelle has developed a radiation computation scheme for use in the Spalding type of recirculating flow code. These efforts are useful for providing insight into the effects of radiation on NO_X formation.

Fuel Nitrogen

Fundamental studies of fuel NO_x conversion are in a preliminary state; most useful results in relating fuel NO_x to equipment parameters have derived from fuels R&D[36,39].

Shell[40], in an exploratory review of the fate of fuel bound nitrogen, concluded that most of the nitrogen goes to the char residue and may be converted to NO during heterogeneous carbon burnout. IFRF has suggested, however, that nearly all fuel NO_x is formed from the gaseous pyrolysis products. Rocketdyne[37] is doing a comprehensive study of several aspects of fuel NO_x conversion which is helping to clarify the mechanisms involved.

Both Shell and Rocketdyne have suggested that significant quantities of HCN may be formed as an intermediate compound of the fuel nitrogen. Concentrations of CN-type molecules, possibly HCN, in excess of 30 ppm have been measured in an experimental burner by Appleton and Heywood[41]. This aspect of fuel nitrogen conversion requires attention, as it could be a limitation on the extent to which conversion of fuel nitrogen to NO is suppressed.

Droplet or Particle Combustion and NO Formation

This is a supporting area for fuel nitrogen and luminous radiation studies and has benefited from non-EPA programs in chemical process engineering. There are numerous models for the combustion of single oil droplets or coal particles; recently several groups[70 - 73] have presented models for NO formation around oil droplets. The Rocketdyne study on fuel nitrogen will include two-phase flow modeling as part of their program.

5.4 APPLICATION OF COMBUSTION MODIFICATIONS

As discussed in the Esso report[1] and in the NO_x control technology literature subsequent to it, combustion modification is the most viable means of reducing NO_x generation. This section presents a review of the implementation strategy, the present status and prospects, and the costs of implementing the various modifications.

5.4.1 Implementation Strategy of Combustion Modifications

As discussed in Section 5.1, NO formation in flames depends on combustion physics and chemistry; specifically, time, temperature, and stoichiometry. Understanding and then altering these fundamental processes is the basis for the effectiveness of combustion modification techniques.

This subsection briefly discusses the theory and implementation strategy of these modifications. Both operating condition and equipment modifications are covered. The bulk of this discussion applies to utility and industrial boilers.

5.4.1.1 Low Excess Air Operation (LEA)

Low excess air firing in boilers reduces the concentration of oxygen available for combination with nitrogen (atmospheric or fuel-bound), thus reducing NO_x formation. Since a certain amount of excess air is always required in practical combustion systems, LEA operation is capable of causing poor combustion with resultant production of unburned fuel and smoke. On oil and coal firing, furnace slagging is also increased with decrease in excess air causing increased maintenance and possible operating problems. Implementation of LEA requires minimal additonal operating expense, no equipment alterations or redesign, and usually results in a slight increase in thermal efficiency.

At the present time, most utility boilers operate at what is considered minimum excess air, and LEA is not considered a combustion modification in the true sense of the word. The minimum level is set by CO levels for gas and smoke levels for oil. Oil requires 1/2 - 1 percent more excess air or O_2 than gas.

The Esso[1] report presented some early results on the effectiveness of LEA operation on gas- and oil-fired boilers. At the time, little or no information was available on the application of LEA operation to coal-fired units. Progress since then is presented in later sections.

5.4.1.2 Off-Stoichiometric (O/S) Combustion

This NO_x reduction technique is usually implemented on large boilers with multiple burners arranged in rectangular matrices mounted either on one boiler wall (front-fired) or on opposite walls (horizontally-opposed-fired). This method can also be used on corner-fired boilers (tangentially-fired), but in most cases the normally low NO_x emissions from boilers of this type can be adequately reduced by simpler techniques, such as lowering excess air.

In general, this method has the effect of fuel-rich burner operation, already identified as an effective NO reducing process, with combustion of the rising bulk gases in lower temperature post-flame regimes where the remainder of the required air is introduced. The time duration for which most of the fuel is exposed to peak temperatures is, therefore, reduced. Depending on the stoichiometry, fuel-rich primary zone combustion may have the secondary effect of lowering the peak flame temperature.

In off-stoichiometric firing, the flame is long, yellow, and smoky, as opposed to the short and intense flame observed on normal firing. The flame also extends farther up in the furnace, sometimes causing excessive reheat (convective section) temperatures. On some units, increased operator vigilance is required due to decreased effectiveness of the flame detector system.

In practice, off-stoichiometric combustion consists of operating some burners (usually the ones located in the lower part of the pattern) fuel-rich while the burners in the upper part of the pattern operate on pure air. Off-stoichiometric combustion is a generic term and several modes of operation are associated with it.

So-called "simulated overfire air" operation results when the top row of burners operate on pure air. "Two-stage" combustion works on the principles of off-stoichiometric combustion except that the fuel-rich burner operation is achieved by diverting a portion of the total required air through separate ports located above the burner pattern. This is also known as "overfire air/NO_x port" operation. In certain boilers, NO reduction optimization requires that the burners operate either fuel- or air-rich in a staggered configuration. This is sometimes called "biased" fired.

The "two-stage" combustion technique is shown in Figure 5-4 below.

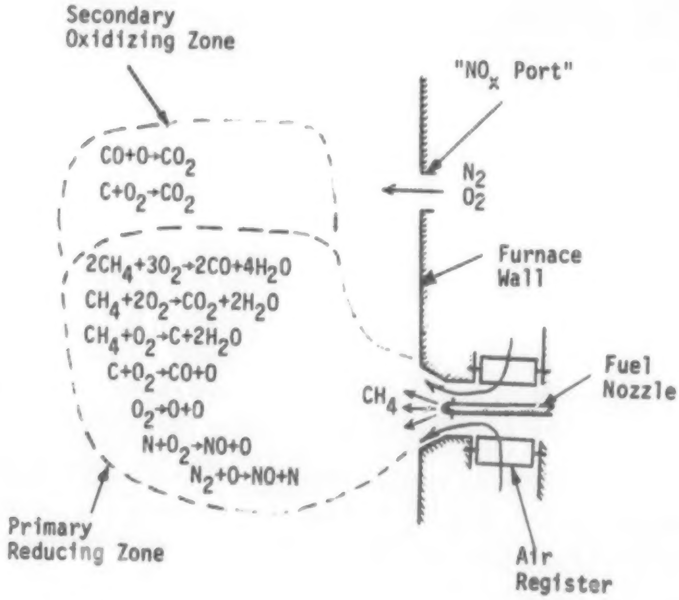

Secondary Oxidizing Zone
"NO_x Port"
$CO+O \rightarrow CO_2$
$C+O_2 \rightarrow CO_2$
N_2
O_2
Furnace Wall
$2CH_4+3O_2 \rightarrow 2CO+4H_2O$
$CH_4+2O_2 \rightarrow CO_2+2H_2O$
$CH_4+O_2 \rightarrow C+2H_2O$
$C+O_2 \rightarrow CO+O$
$O_2 \rightarrow O+O$
$N+O_2 \rightarrow NO+O$
$N_2+O \rightarrow NO+N$
CH_4
Fuel Nozzle
Primary Reducing Zone
Air Register

Figure 5-4. Two-Stage Combustion (After Reference 35)

A vertical cross-section of a utility boiler burner is shown schematically. Two-stage combustion of natural gas (methane) is depicted, and a few of the global reaction mechanisms associated with the primary and secondary combustion zones are identified.

As of 1969, the date of publication of the Esso[1] report, only a limited quantity of information was available on the actual effectiveness of off-stoichiometric techniques for NO reduction in boilers. Fairly significant results had been obtained for gas-fired utility boilers by Pacific Gas and Electric Company and Southern California Edison Company, and from coal-fired, sub-scale combustion in tests performed by the U.S. Bureau of Mines (1966). This modification technique has been more thoroughly investigated during the last several years, and subsequent sections of the present study review the recent developments.

5.4.1.3 Flue Gas Recirculation (FGR)

A portion of the flue gas recycled back to the primary combustion zone reduces NO formation by acting as a thermal ballast to dilute the reactants, thereby reducing both the peak flame temperature and partial pressure of available oxygen at the burner inlet. Possible flame instability, loss of heat exchanger efficiency, and, for packaged boilers, condensation on internal heat transfer surfaces, limits the utility of FGR on some units.

Although it has been concluded that FGR reduces thermal NO, recent data has cast doubts on its capability to reduce fuel NO. More investigation is required, as the present uncertainty will have significant impact on the applicability of FGR to oil and coal firing.

At the time of the Esso report there existed a paucity of information concerning the effectiveness of this method on NO reduction, the only reported work being performed by Andrews, et. al.[74]. New data has come to light since then, and is included in Section 5.4.2.

5.4.1.4 Reduced Air Preheat Operation

Reducing the amount of combustion air preheat lowers the primary combustion zone peak temperature, generally lowering NO production as a result. It is seldom considered practical, however, due to the associated loss in thermal efficiency.

There have been recent indications that increasing air preheat actually reduces fuel NO_x formation on coal firing[92]. Further investigations are warranted to explain this result.

This modification technique was not mentioned in the Esso[1] report due to a lack of empirical data concerning its effectiveness. It may have some merit, however, and will be discussed further in Section 5.4.2.

5.4.1.5 Load Reduction

The term "load" is defined as the percentage of its rated capacity at which the furnace or boiler is being operated. Increasing boiler load causes an increase in primary combustion zone volumetric heat release rate which generally increases the rate of NO formation. Reducing boiler load is accomplished by reducing the reactant flow rate (fuel and oxidizer) into the furnace, thereby lowering the heat release rate (also known as combustion intensity) and peak flame temperature.

Although there are no capital costs involved in implementing load reduction, it is usually undesirable to reduce boiler capacity. Load reduction increases boiler efficiency due to longer residence times and more complete combustion, but the lower flow rates result in a decrease in overall efficiency due to decreased turbine efficiency.

This modification technique was not mentioned in the Esso report due to a lack of empirical data concerning its effectiveness. It has some merit, however, and is briefly discussed in Section 5.4.2.

5.4.1.6 Water or Steam Injection

Injecting water or steam into boilers is yet another means of reducing peak combustion temperatures to reduce NO formation. Its use may entail some undesirable operating conditions, such as decreased thermal efficiency due to the high heat capacity of water compared with that of flue gas or other inert diluents, and increased equipment corrosion.

The Esso report noted from one study that water injection was effective in reducing NO_x emissions from an internal combustion engine. No other investigation had been carried out up to that time. It has since been shown to be a viable means of NO_x suppression for stationary gas turbines.

5.4.1.7 Combinations of Techniques

Since 1969, it has been demonstrated that several of the previously discussed modification techniques can be effectively utilized in combination since they reduce NO by different mechanisms. Most often, off-stoichiometric combustion is used in conjunction with flue gas recirculation, low excess air, reduced air preheat, or reduced load. The latter three methods lower peak combustion temperatures, while off-stoichiometric operation reduces the amount of fuel burned at peak temperature. For the most part, combining techniques has been shown to be complementary but not additive for NO reduction.

5.4.1.8 Equipment Design Modification

From what is known of the theory of NO_x formation in continuous combustion processes, the ideal equipment design would provide for lower peak temperature flames and more controlled rates of combustion. Research and testing has shown that much can be done to accomplish these goals.

According to Krippene[75] a low NO_x emission burner should be designed as fuel-flexible as possible, be able to operate at minimum excess air levels, and not cause an emissions tradeoff problem. In most new utility boiler designs, burner spacing has widened to provide more cooling of the burner zone area. In addition, the furnace enclosure should be built to allow sufficient time for complete fuel combustion from slower and more controlled heat release rates, such as that associated with the off-stoichiometric operating mode.

As mentioned in the Esso report, the tangential firing method for large utility boilers is in itself a viable low NO_x emission technique. Notable results have been obtained from tests of tangentially-fired units, by Combustion Engineering[76 - 78]. It has been observed, however, that from many such boilers the response to operating modifications has been less impressive than from boilers of other designs, even though the magnitude of the initial, uncontrolled emission was lower.

5.4.1.9 Typical Utility Boiler Combustion Modification Implementation Program

As a vehicle for emphasizing the importance of cost-benefit factors, the Esso report[1] contained an extensive analysis of minimum cost paths for NO_x control by combustion modification. An updated study of this kind is required, since the original is based on estimated costs and the fuel NO_x problem was not fully appreciated at the time. An approach for beginning such an analysis is presented in Figure 5-5.

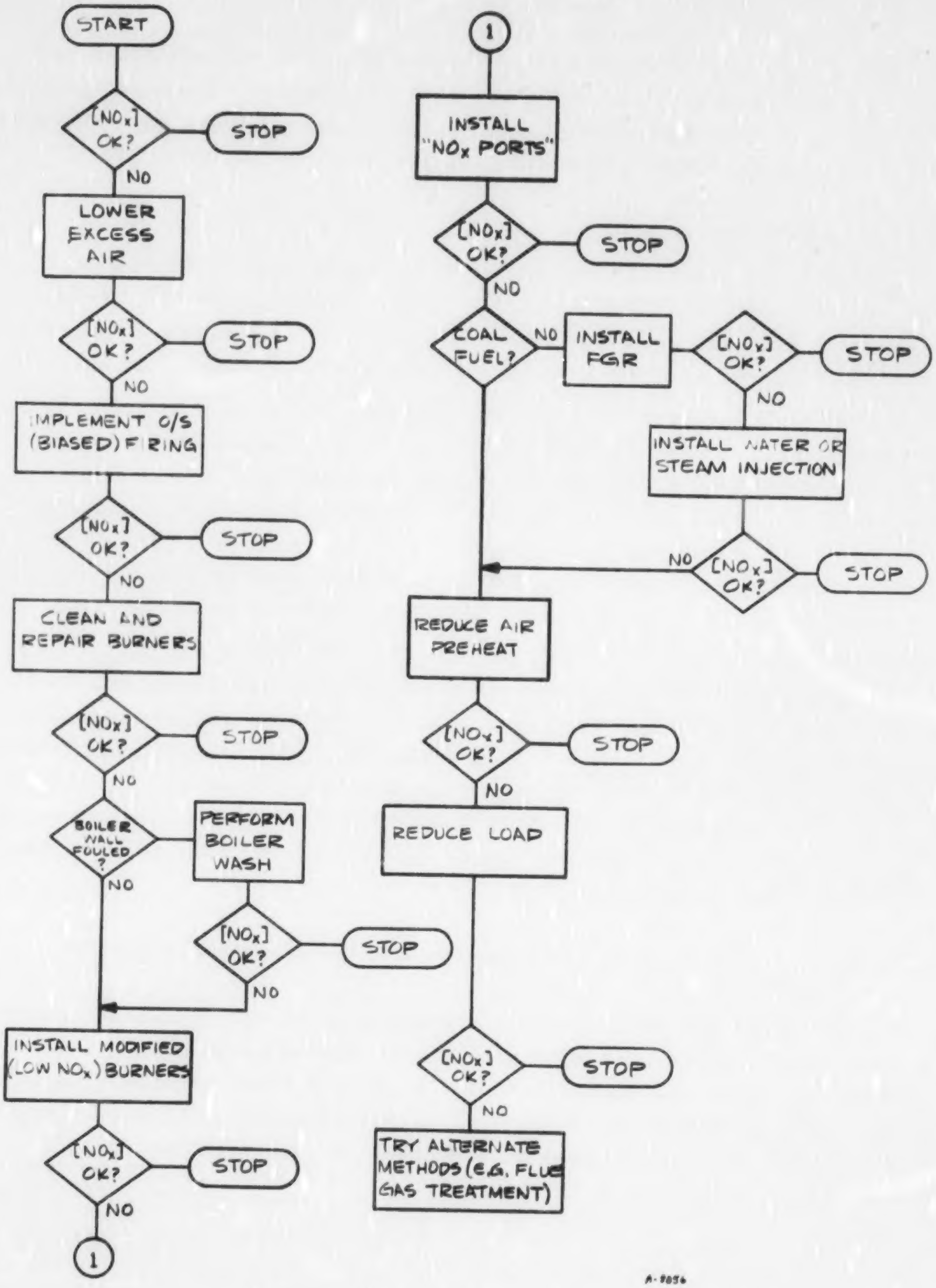

START
[NOx] OK?
STOP
NO
LOWER EXCESS AIR
[NOx] OK?
STOP
NO
IMPLEMENT O/S (BIASED) FIRING
[NOx] OK?
STOP
NO
CLEAN AND REPAIR BURNERS
[NOx] OK?
STOP
NO
BOILER WALL FOULED?
PERFORM BOILER WASH
NO
[NOx] OK?
STOP
NO
INSTALL MODIFIED (LOW NOx) BURNERS
[NOx] OK?
STOP
NO
1
1
INSTALL "NOx PORTS"
[NOx] OK?
STOP
NO
COAL FUEL?
NO
INSTALL FGR
[NOx] OK?
STOP
NO
INSTALL WATER OR STEAM INJECTION
NO
[NOx] OK?
STOP
REDUCE AIR PREHEAT
[NOx] OK?
STOP
NO
REDUCE LOAD
[NOx] OK?
STOP
NO
TRY ALTERNATE METHODS (E.G. FLUE GAS TREATMENT)
A-9856

Figure 5-5. Typical Combustion Modification Implementation Program

Figure 5-5 shows, in flow-chart form, a typical decision process for implementation of the appropriate combustion modification techniques. The simplest, least expensive, and most expedient techniques are initially utilized, followed by those requiring capital investment, operating expense, boiler down-time, and decreased thermal efficiency. This simplistic model reflects the possible ineffectiveness of temperature-reduction schemes (i.e., FGR, water injection) on emissions from coal combustion, and has as its ultimate goal the reduction of NO concentration (shown as [NO_x]) to regulation levels. In practice, obviously, the decision process is more complex. Impracticalities of certain of the techniques for a given boiler may require an extensive re-ordering of the control steps as given in the figure.

5.4.2 Status and Prospects

Methods of controlling NO_x emissions from combustion systems by combustion modification techniques were treated at some length in the Esso report. Since 1969, however, much more experience in testing and applying these operating modes has been gained.

This section discusses the present status and prospects for reducing NO_x emissions from stationary sources by combustion modifications. The scope of this section focuses on those combustion systems usually categorized as "point" sources: utility and large industrial boilers. "Area" or "complex" sources, comprised partly of stationary I.C. engines and gas turbines, are also discussed. Other area sources, such as commercial and domestic heating equipment, are discussed when similarities exist with point sources.

5.4.2.1 Utility and Industrial Boilers

The net decrease in NO_x emissions from conventional fossil-fueled boilers through changes in operating mode or equipment design depends on many factors. Among them are boiler geometry and cleanliness, burner design, spacing and state of repair, load, and most importantly, the fuel itself. In this section, the relative effectiveness of the various combustion modification techniques for reducing NO from gas-, oil-, and coal-fired boilers or combustors will be discussed. This information was obtained from the open literature and a number of confirmatory personal contacts.

When attempting to draw general conclusions from NO_x reduction figures obtained from separate boilers, it must be kept in mind that even identical boilers give widely differing test results for the same modification. This is due to varying boiler cleanliness, uneven burner loading, equipment condition, and a myriad of other factors, most of them not easily identifiable. Unless the sample size is large, extrapolations based on boiler size are probably invalid.

As implied in Figure 5-5 in the preceding section, the practical limit on the modifications are based initially on three subjective criteria: stack plume appearance (i.e., smoke production, its opaqueness and/or Ringlemann number), flame appearance (i.e., color and dimensions) and incipience of flame instability at the burner. When problems are encountered, implementation is halted and the situation re-evaluated. Stack gas sampling for NO_x, CO, and O_2 is usually carried out concurrently. In the long term, the effects of the modification on such factors as burner condition, furnace slagging and corrosion, ability to change fuels, and boiler load are monitored to varying degrees.

5.4.2.1.1 Gas

The highest degree of success in reducing NO by the application of combustion modifications has been obtained on gas firing for both full and pilot scale furnaces. The reason for this effectiveness lies in the fact that all of these techniques reduce thermal NO_x, which is the only NO_x formation mechanism in gas combustion.

Low excess air operation has been shown to be extremely effective in lowering NO_x emission from gas-fired boilers. An extensive study of NO_x reduction techniques applied to six wall-fired utility boilers done by Bartok, et al.[79] showed reductions of 25 to 60 percent at full load. NO_x reduction magnitude depended not only on lowering excess air, but also on furnace design and firing method.

In other reports, Barr[80] obtained a 23 percent NO reduction on a 750 MW front-fired unit as a result of lowering excess air. Off-stoichiometric firing was subsequently implemented on this unit to achieve further reductions. Blakeslee[77] reported a 33 percent NO_x reduction on a 250 MW tangentially-fired utility boiler when the flue gas oxygen content was decreased from 3.9 percent to 0.6 percent.

In most cases, LEA was implemented without serious flame stability problems, and an increase in thermal efficiency was noted.

From both full and pilot scale results, flue gas recirculation has been proven effective for lowering NO_x formation from gas combustion. In general, NO_x reduction figures range from 20 to 60 percent on various boiler designs and load conditions. Subscale testing has shown that the magnitude of NO_x reduction is mainly dependent on the amount of gas recirculated up to the point of incipient flame instability and other undesirable operating conditions. Some references which may be scanned for further details are References 79, 82, 83, and 78.

On gas firing, off-stoichiometric combustion has been shown to be one of the most effective means of NO_x control and also one of the easiest to implement. Biased firing is the most frequently used and most effective method, while overfire air/NO port operation achieves less reduction, particularly where biasing has already been implemented. Bartok[79] published NO_x reduction figures of 25 to 58 percent from wall-fired utility boilers ranging from 80 to 480 MW when two-stage combustion was applied. Similar results are reported on Southern California Edison units by Bagwell[82], Breen[84], and Teixeira[85]. The effectiveness of off-stoichiometric combustion on gas-firing has been well researched and documented.

Water injection into utility boilers has been tested to a limited extent. Blakeslee[77] reports a 50 percent maximum NO reduction at full load for a 250 MW tangentially-fired unit when water was injected at a rate of 45 pounds per million Btu fired. Boiler convective section temperature increased by 250°F and boiler efficiency dropped 5 percent. The economic penalties resulting from this method, as well as from reduced load or air preheat, make such techniques unattractive.

A significant amount of work has been done on optimizing gas burner design for low NO_x. Of the three types of burners, spuds, radial spud, and ring, the latter forms the least NO_x while the spud type maximize NO_x formation. In addition, burners which produce low turbulence flames have been found to release lower quantities of NO_x.

In all, the effectiveness of NO_x reduction from most gas-fired equipment has been adequately explored. Further investigation may not be warranted because the number of large gas-fired utility and industrial boilers (the largest point NO_x sources), small to begin with, is now declining rapidly due to the present natural gas supply shortage. For example, several West Coast utilities estimate reductions from about 70 percent gas-firing in 1972, to less than 10 percent in 1974. Low sulfur residual oil will be the predominant fuel utilized on the West Coast.

5.4.2.1.2 Oil

In general, a poorer record of NO_X reduction has been compiled for oil-fired units, largely because of reduced operating flexibility. Fuel NO_X becomes an important contribution to the total NO_X emission from a given unit, and the individual modifications are less effective and more complicated to implement. Nevertheless, substantial reductions have been achieved, in some instances as high as 50 percent on utility boilers.

As for gas, operating at minimum excess air conditions is a valid reduction scheme. From tests on six wall-, oil-fired utility boilers, Bartok, _et_ _al_.[79] reported a NO_X reduction range of 23 - 56 percent at full load. LEA operation seemed most effective on front wall-fired units, although this sample size was rather small.

On other utility boiler tests, Campbell[86] reported a 17 percent NO_X decrease by lowering excess O_2 from 3.8 percent to 1.5 percent on a 290 MW front-fired unit.

No reliable correlations between furnace design and effectiveness of LEA operation on utility boilers have been or can be made at this time.

The importance of avoiding smoke production when approaching reducing conditions at the burner has been well documented. A typical example was generated from sub-scale testing by Muzio and Wilson[87] on a 3.7 x 10^6 Btu/hr research furnace. Figure 5-6 depicts the NO and smoke trends as a function of excess air.

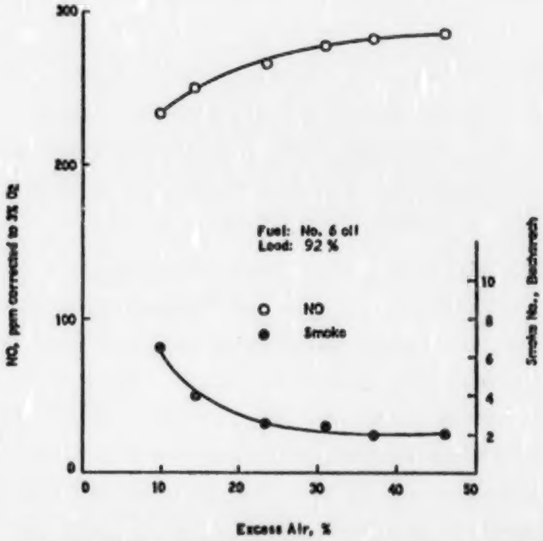


Figure 5-6. Increasing Excess Air Decreases Smoke but Increases NO

The thrust of other recent subscale testing has been to segregate the relative importances of thermal and fuel NO under low excess air conditions. Firing "blank" fuels (i.e., no nitrogen) doped with varying amounts of nitrogen-bearing compounds, Turner[88] found that at any given level of excess air, the NO_X concentration increases with fuel nitrogen content, although not proportionately. As excess air was increased, the fraction of fuel nitrogen converted to NO increased, but the fraction decreased with increasing nitrogen content at a given excess air level. Total NO concentration in the flue gas, however, increased with fuel nitrogen concentration. These conclusions have been substantiated by Pershing[89]. In a more recent study, Martin[90] found that although fuel NO decreased as excess air was lowered, thermal NO actually increased. This behavior was attributed to specific characteristics of the furnace used in the investigation. It was concluded, however, that limiting excess air may not, a priori, be a valid control technique for all small oil-fired boilers.

According to Jain[81], commercial and industrial boilers generally operate at an average excess air level of 15 to 25 percent, with some smaller and older units operating at as high as 35 to 40 percent excess air. Early tests have shown that these levels can be safely reduced to about 10 percent, causing substantially lower NO_X emissions. As for gas-fired boilers of this size range, a number of field testing and survey programs are currently being performed to more fully characterize the NO_X contributions from small oil-fired boilers.

Off-stoichiometric combustion has also been demonstrated to be effective in reducing NO_X from oil-fired power plants. Bartok, et al.[79], reported net NO_X reductions of 19 to 35 percent for five wall-fired boilers ranging in size from 80 to 320 MW. Blakeslee[77,78], obtained reductions of 20 to 55 percent for several 78 to 400 MW tangentially-fired units when two-stage combustion was implemented. Similar impressive results were reported by Teixeira[85] from the NO_X reduction program being carried out at Southern California Edison. Some research has shown that off-stoichiometric techniques reduce both fuel and thermal NO_X formation. Such conclusions were arrived at by Turner[88] in doped fuel tests.

Although more work, especially full-scale field testing, is required to investigate the effectiveness of off-stoichiometric methods on reducing fuel NO_X, the effectiveness of these techniques on net NO_X emissions from large boilers has been solidly demonstrated. Smaller industrial and commercial boilers, however, do not lend themselves to such methods. For packaged boilers, furnace volume and the number of burners are usually too small, and implementation of such modifications is usually uneconomical[10,81].

For flue gas recirculation on full size utility boilers, reduction figures in the range of 20 to 40 percent are most common[78]. Bartok, et al.[79] reported a 50 percent reduction in NO_X emissions from a 250 MW front-fired twin furnace when FGR was combined with low excess air and staged firing operating conditions. Similar results from combining techniques have been obtained by Barr and James[13] from a 750 MW unit at full load.

The importance of introducing the flue gas directly into the combustion air as opposed to injection into the furnace volume is described in a paper by Campbell[86]. On a 342 MW oil-fired unit, recirculating gas from the economizer outlet to the upper furnace area caused no change in NO_X , while on a 250 MW oil-fired utility boiler, routing the gas through the windbox produced a 15 percent NO_X reduction at 80 percent load.

Sub-scale research[39,88 - 90] has shown that flue gas recirculation with liquid fuel combustion reduces NO_X formation by the thermal fixation mechanism but does not affect fuel nitrogen conversion. Consequently, less significant total NO_X reductions would be expected when FGR is applied to units firing fuels bearing nitrogen compounds. Typical results in this regard are shown in Figure 5-7, from work done by Muzio and Wilson[87] on a 3.7 x 10^6 Btu/hr research furnace. The role of fuel nitrogen conversion is clearly stated when comparing the curves for natural gas and nitrogen-containing No. 2 and No. 6 fuel oils.

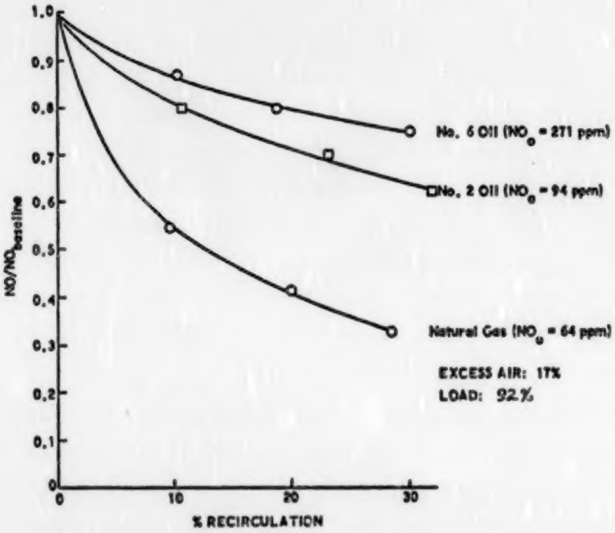


Figure 5-7. Effectiveness of FGR with Fuel Oil Type

Although it is estimated that substantial reductions may be obtained by FGR on commercial and industrial boilers, essentially no published data exists to back up this claim. The Esso report discussed the early work done by Andrews, et al.[74], and this seems to be the state of the art. In any case, installation of FGR on existing package boilers would require capital investment in ducts, fans, and control systems, as well as additional operating costs due to the power needed for a forced draft system. Generally applicable to typical package boiler emission control methods is the research reported in Reference 87.

Water injection is currently being tested on one unit in Southern California. Due to the loss in plant efficiency usually assoicated with this method, it may be used to a limited extent in combinations with other techniques to supplement their NO_x reduction capabilities.

In the area of burner modifications for oil-fired utilities, one of the most straightforward is installing new atomizing orifices for biased firing. For those burners operating fuel-rich, the orifices are enlarged to accomodate the increased fuel flow caused by the burners out of service.

Krippene[75] states the criteria for a low NO_x burner. The design should afford a limited turbulence, controlled diffusion flame, with a combustion process reminiscent of off-stoichiometric techniques. The quantities of fuel and air mixed at the burners should be minimized to that required to sustain combustion, while the remainder of the fuel is combusted slowly and efficiently further downstream. Burner flexibility is crucial for maintaining efficient combustion of a wide range of fuel oils under varying load conditions.

In the near future, oil will become an increasingly important fuel for stationary sources. Fortunately, many of the installations that will soon be converted from gas- to oil-firing were originally designed for dual fuels. Many more, however, will require some degree of modification before compatibility with oil is attained. Furthermore, such units must retain the highest degree of flexibility possible to assure efficient combustion of a wide variety of fuel oils at consistently high load levels.

5.4.2.1.3 Coal

Just as the response to combustion modifications for oil-firing was less impressive than for gas, so their effectiveness for coal fuel is diminished further. The contribution from fuel N conversion is very important and the number of cost-effective modifications is commensurately reduced. Since the possible number of operating problems on coal-firing are more numerous and the propensity to produce smoke or particulates is greater, the modifications which are effective are less flexible and must, in all cases, be applied with great care. Most of the existing data on this topic has been generated from pilot-scale tests, with a limited amount from large utility boilers. KVB Engineering of Tustin, California, is currently conducting an EPA-sponsored industrial boiler NO_X control study. Due to the negligible amount of presently available information on coal-fired commercial and industrial boilers, the following discussion focuses on utility boilers.

In tests on eight utility boilers ranging in size from 125 to 800 MW, Crawford[91] reported average NO_X reduction figures of 40 to 50 percent under low excess air and biased firing conditions. Tangentially-fired units produced the lowest NO_X under both baseline and modified operation, and load was not effected during burner pattern firing changes for wall-fired units. Increased slagging was a problem, and it was recommended that long term corrosion investigations be made on future coal testing.

The dependence of net NO_X reduction on fuel type for given modification variables was shown in sub-scale tests (5.6×10^6 Btu/hr, 500 lb/hr coal-fired furnace) by Armento and Sage[92]. Figures 5-8, 5-9 and 5-10 show NO_X emission as a function of excess air, firing rate (load), and preheat, respectively.

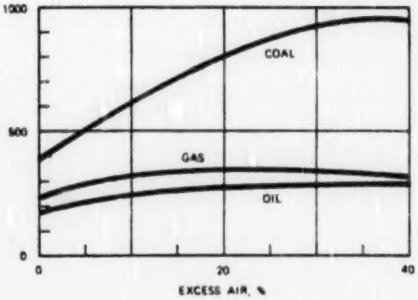


Figure 5-8. Effect of Excess Air on NO_X for Gas, Oil, and Coal

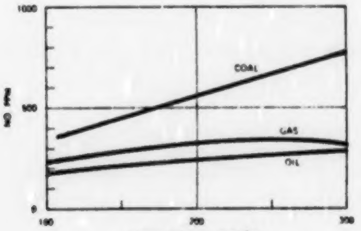


Figure 5-9. Effect of Load on NO_X for Gas, Oil, and Coal

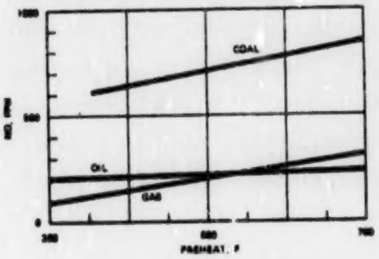


Figure 5-10. Effect of Preheat on NO_x for Gas, Oil, and Coal

It was concluded that coal produced more NO_x for all modifications due to fuel N conversion. For this particular experimental system the excess air and load load tests showed that oil produced less NO_x than gas. These results were due to the more luminous and larger oil flame envelope, leading to greater radiation and a lower bulk gas temperature. Except for this phenomenon, the authors concluded that the observed trends shown in Figures 5-8 and 5-9 would be valid and useful scale-up information.

As was stated earlier in this report, there is some conflict over whether or not temperature reduction schemes, such as flue gas recirculation and water injection, are effective for reducing NO_x on coal- or heavy oil-firing. The sub-scale testing by Armento and Sage produced a 10 to 15 percent maximum reduction in NO_x for the application of 10 to 15 percent flue gas recirculation, as shown in Figure 5-11. The oil fired in the tests contained 0.23 percent fuel-bound nitrogen and the coal 1.1 percent. Again, it was proposed that the more luminous oil flame caused FGR to be less effective on oil than on coal, just the opposite of what is normally expected.

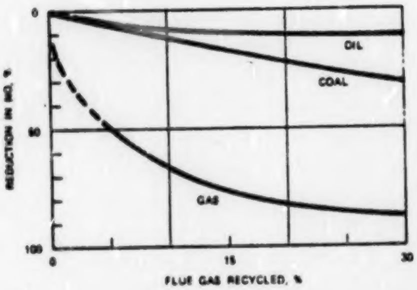


Figure 5-11. Effect of FGR on NO_x for Gas, Oil, and Coal

A wide variety of results have been published by other investigators. McCann, et at.[93], reported a 39 percent NO_x reduction at 25 percent flue gas recirculated to secondary air for a 500 lb/hr pulverized coal-fired unit. These results may be questionable, since the concurrent carbon loss increase data showed that incomplete combustion was taking place. From tests by Pershing, et al.[89] on a 4 lb/hr pulverized coal-fired furnace, it was concluded that FGR was ineffective as a NO_x reduction technique.

Most researchers have agreed that decreases in NO_x on coal firing by FGR were relatively minor and were probably associated with the elimination of thermal NO_x. The larger portion of the total NO_x results from conversion of fuel-bound nitrogen. FGR would perhaps be used to supplement the reductions obtained by other techniques, such as off-stoichiometric combustion, that are postulated to be effective for fuel NO_x reduction.

According to Krippene[75], much can be done to alleviate operational problems through combustion equipment modification. To provide closer control over fuel and air flow through the burners, some coal-fired utility boilers have installed individual windboxes for groups of burners supplied by a given pulverizer. This is an important modification for biased firing since continuous air purge through the burners out of service is a requirement.

The "low NO_x" furner described in the preceding section on oil firing is also effective on coal firing. Defined as a "triple concentric" burner due to its unique design, it produced a combustion process similar to the two-stage technique used on a boiler as a whole.

As discussed in Sections 5.1 and 5.2, research has shown that there are possibly two different fates in store for fuel-bound nitrogen during and after combustion. It will either be converted to N_2, or NO, along with other intermediate products (several of which have been identified as cyanide compounds). These reactions depend more on stoichiometry than on temperature. Under oxidizing conditions more NO is produced, while nitrogen is the favored product from a reducing atmosphere, such as that afforded by off-stoichiometric combustion. In full- and sub-scale tests so far, these techniques have been shown to be quite effective for reducing NO_x on coal-firing, and will probably be implemented on new and existing coal-fired utility boilers and on boilers being converted from scarcer fuels to coal.

As discussed in Section 3.4 of this report, the current clean fuels shortage and slower-than-anticipated construction of nuclear generating plants has vaulted coal, the use of which had been steadily declining over the past 30 years, into prominence as a major short-term energy resource. To avoid any further degradation in ambient air quality, emissions from the burgeoning number of coal-fired stationary sources must be controlled.

Specifically for NO_X abatement, this will require further pilot- and full-scale research and development on the NO_X reduction effectiveness of combustion equipment and operation modifications with minimum economic penalties.

5.4.2.2 Turbines and Reciprocating Internal Combustion Engines

This section updates the status of the control technology for NO_X emissions from stationary reciprocating I.C. engines and gas turbines.

5.4.2.2.1 Reciprocating I.C. Engines

Most of the control technology for stationary I.C. engines has been developed by the automotive industry. Table 5-4 lists the control techniques for all exhaust pollutants.

TABLE 5-4

EMISSION CONTROL METHODS FOR RECIPROCATING ENGINES

			Discussed in Esso Study
I. Engine Modifications			
	A. Operating Conditions:	1. Speed	
		2. Torque/Load	
		3. Air/Fuel Ratio	X
		4. Ignition Timing	X
		5. Fuel Injection Timing	X
		6. Air Temperature	
		7. Air Pressure	
		8. Exhaust Back Pressure	
	B. Engine Hardware:	1. Exhaust Recirculation	X
		2. Water Injection	X
		3. Valve Timing	
		4. Combustion Chamber - Stratified Charge	X
		5. Compression Ratio	X
II. Exhaust Treatment			
	A. Exhaust Thermal Reactor (CO/HC)		
	B. Catalytic Converter:	1. Oxidation (CO/HC)	
		2. Reduction of NO_X by CO, H_2, NH_3, or natural gas	

In general, changes that reduce NO_x emissions have the reverse effect on CO and hydrocarbon emissions and fuel consumption.

The optimum choice for any particular engine will depend on the effect of the controls, reliability, durability and engine life. A study by McGowin[4] recommended the following NO_x control techniques for stationary diesel and natural gas engines:

ENGINE	SHORT & IMMEDIATE TERM	LONG TERM
Diesel	Water Injection Precombustion Chamber	Catalytic NO_x Reduction
Natural Gas	Water Injection Increased valve overlap for 4 cycle N.A. engines	Catalytic NO_x Reduction

The precombustion chamber or stratified charge engine has the potential of reducing diesel NO_x emissions by a factor of two. NO_x emissions can also be reduced by 70 percent or more by injection of water into the combustion chamber at the rate of 1 lb of water for each pound of fuel. However, fuel consumption is likely to increase and the long term effects of this practice are not known. The parameters or control techniques that were omitted in the Esso study or schemes where significant new data have been developed will now be discussed.

Engine Speed

For a 4-cycle gasoline engine, NO_x emission increases with speed under fuel-rich conditions, decrease with speed under fuel-lean conditions and remain nearly constant at the optimum air/fuel ratio. The Shell study[4] showed a 58 percent decrease in NO_x emissions with a 10 percent increase in speed for a two cycle, naturally-aspirated gas engine. The decrease may be partly attributable to simultaneous change in the apparent air/fuel ratio in the cylinder.

Torque Load

The Shell study[4] also showed a dramatic increase in NO_x emission with increasing torque. Emissions increased from 15 gms/Bhp-hr to 23 gms/Bhp-hr when increasing the torque from 83 Bmep to 90 Bmep on a two stroke, atmospheric spark-gas engine. This effect is further illustrated by data developed at Caterpiller Tractor Company for a precombustion chamber diesel engine as shown in Figure 5-12. However, with this type of engine, NO_x emission increase with speed at constant power output.

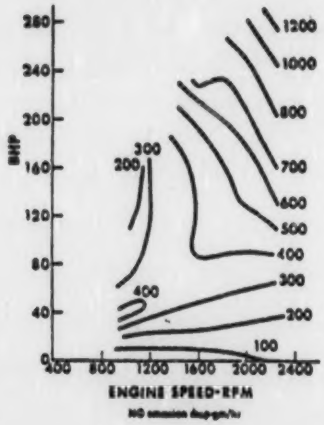


Figure 5-12. Effect of Speed and Power Output on Emissions; Caterpiller 4-Cycle Precombustion Chamber; Diesel Engine

Air Manifold Temperature and Exhaust Back Pressure

As might be expected, NO_x emissions increase with increasing air manifold temperature. For a spark ignition gasoline engine under lean conditions increasing manifold air pressure increased emission. Conversely, increasing exhaust back pressure on a 2-stroke, naturally-aspirated engine decreases NO_x[4].

Valve Overlap

For a four cycle naturally-aspirated engine, increasing valve overlap produces the same effect as exhaust gas recirculation. As the valve overlap increases, the percentage of exhaust present in the fresh charge increases which results in an EGR effect. Fuel economy may suffer, however. Increasing valve overlap can only be applied to four cycle engines.

Exhaust Gas Recirculation

Although this technique was mentioned in the 1969 Esso study, considerable work has been performed since that time. In a recent paper by Komiyama and Heywood[94] a model was developed to predict NO_x emission, given the design and operating conditions for spark ignition engines and percent of exhaust gas recirculation. Good agreement was shown between the model and experimental data over a wide range of fuel/air ratios and engine speeds. Typical reductions of 38 percent have been achieved under lean conditions. The paper shows that in lean mixtures nitric oxide concentration freezes early in the expansion stroke. Under rich conditions considerable nitric oxide decomposition occurs during combustion before the concentrations are fixed. Caterpillar has published EGR data for a precombustion chamber diesel which showed a possible 73 percent NO_x reduction. However, a number of problems exist for EGR applied to stationary I.C. engines, which include the following:

- Requirement to accurately meter the amount recirculated
- Must cool the exhaust products prior to injection
- Fouling of intake manifolds, and aftercoolers by particulates
- Long term effects on lubricating oil and engine life.

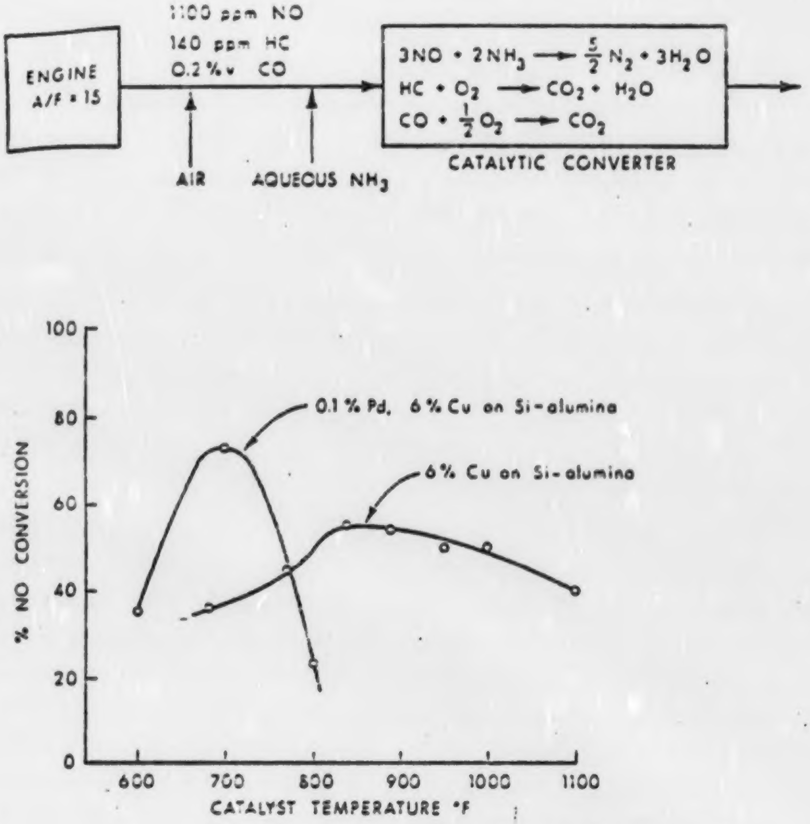
Catalytic Converter

Numerous post-combustion control methods have been developed by the automotive industry to treat the exhaust products from an I.C. engine. These techniques include exhaust manifold thermal reactors, catalytic converters, exhaust gas scrubbers and solid sorbents. The exhaust thermal reactor requires a rich mixture operation and reduces only HC and CO. Exhaust gas scrubbing and solid sorbents create secondary pollution problems, are too costly for single engine application, and there is no known NO_x scrubbing technique. Thus, only the catalytic converter seems to be practical for stationary I.C. engines. Catalytic converters can be designed for the oxidation of CO and hydrocarbons, or reduction of NO_x by CO, H_2, NH_3 or natural gas. Only the latter scheme will be discussed.

NO_x can be removed by catalytic reduction using CO and hydrogen, or by adding a reducing agent such as ammonia or natural gas. Only under rich operating conditions will sufficient hydrogen and CO be present to achieve the required reactions. Since most stationary engines operate under fuel-lean conditions only the addition of natural gas or ammonia seem practical. Injection of natural gas requires that there be sufficient gas to react with the remaining O_2 before NO_x can be reduced. This technique may require several stages of reactors. Of these catalytic reduction methods, the ammonia scheme holds the most promise.

The ammonia reduction technique is shown schematically in Figure 5-13, together with typical conversion curves for two different catalysts. Several advantages of this scheme are:

- Reduction of NO_x takes place in the presence of O_2.
- CO and H/C oxidation occurs simultaneously on the same catalyst.



(REFERENCE; U. S. PATENT 3,449,063)

Figure 5-13. Catalytic Reduction of NO_x by Ammonia

- Allows operation of the engine at maximum fuel economy or power.

Areas of additional R&D work required for system optimization are as follows:

- Determine optimum catalyst composition.
- Determine catalyst durability.
- Determine resistance to catalyst poison present in the fuel (Sulfur and metal impurities).

5.4.2.2.2 Gas Turbines

As with reciprocating engines, the most viable control techniques are engine modifications and exhaust treatment. The comments made for exhaust gas treatment also apply to turbines but it must be remembered that more than twice as much air must be handled for an equivalent output. Thus, the exhaust gas treatment schemes do not seem too practical.

The approaches that are usually taken include combustor chamber design modification, water injection, modification of fuel properties, and exhaust gas recirculation. It is generally known that emissions of nitrogen oxides increase with power level and that power is directly related to maximum local combustion temperatures. NO_x emission can also be directly related to combustor inlet temperature and, in turn, to compressor pressure ratio.

A frequently-used technique has been to lower the peak flame temperature by leaning out the primary zone through combustion chamber design. An alternate method presented by Parikh, Sawyer and London[95] was to increase the homogeneity of the primary zone composition by using gaseous or vaporized liquid fuels. Both schemes can create problems in the ability to maintain flame stabilization and lower lean extinction performance. Reducing the residence time of the hot gases will also lead to lower concentrations of NO. Residence time may be decreased by moving the dilution holes upstream or increasing the flow velocity through the primary zone. Typical reduction of 15 percent are not uncommon[94] for lean primary zone combustors.

Injection of water into the combustor primary zone or injection of steam into the combustion air can typically reduce NO_x emission by up to 85 percent[94]. However, large quantities of demineralized water must be handled and engine maintenance is increased.

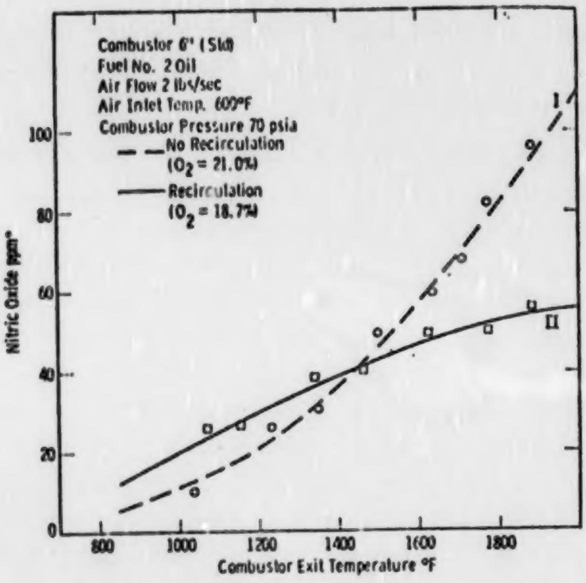
Exhaust gas recirculation has an affect similar to lean primary zone combustion. Typical results for EGR are shown in Figures 5-14 and 5-15 for oil and gas fired systems respectively[96].

NASA has been working on the development of low pollutant-generation combustors ("Clean Combustor Program") for aircraft engines. A great number of concepts are being tested which include lean primary zone combustors, a short residence time combustor, combustion which increases the homogeneity of the primary zone composition through the use of multiple arrays of "swirl-can" modules or a premix package, and two stage combustors which use a "pilot combustion zone" (similar to the precombustion chamber diesel engine). Variable geometry combustors are also being explored to achieve the most favorable configuration for any load. However, a variable geometry scheme will not be necessary for stationary gas turbine installations. Details of these configurations are reported in a paper by R. E. Jones[97]. To data none of the combustor designs have achieved the desired goal for the proposed 1979 EPA limits for aircraft engines but it is felt that some of the designs will have the potential for further NO_x reduction.

5.4.3 Cost

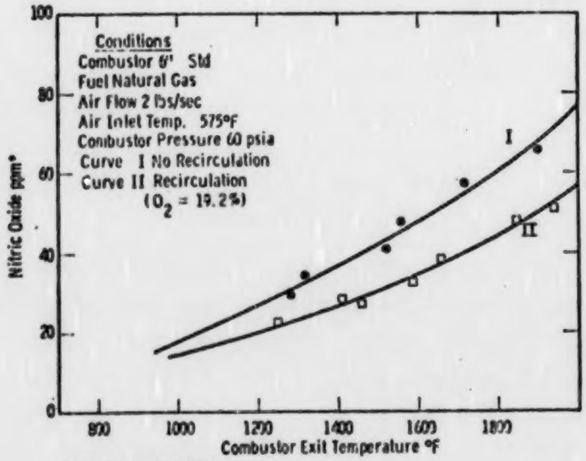
There is an overall paucity of reliable data on the costs involved in implementing combustion modifications on either conventional utility and industrial boilers or stationary gas turbines and reciprocating I.C. engines. Much of the cost information is proprietary, and the overall state of definitive cost analyses is still quite primitive. The data exhibit a wide range of costs for a given modification on the same type of combustion system. In any case, available information will be presented in this section, but without extensive critique or evaluation. The bulk of the discussion will center on utility and industrial boilers. Limited cost data for stationary I.C. engine and gas turbine modifications were published by McGowin[4].

The costs of implementing the combustion modification techniques described in preceding sections is basically the sum of the initial capital cost, annual capital cost, and annual operating cost (which includes any cost savings). Based on estimates for these costs, the Esso report presented the results of a cost effectiveness study performed for NO_x control on utility boilers by means of combustion modification. Since 1969, however, it has been revealed that a wide variation in the effectiveness of the control techniques among boilers exists. This problem will require that future cost-effectiveness evaluations be done on an individual boiler basis.



* Dry Volumetric Basis

Figure 5-14. Effect of Cooled Exhaust Gas Recirculation on NO Emission (Using Oil as Fuel)



* Dry Volumetric Basis

Figure 5-15. Effect of Cooled Exhaust Gas Recirculation on NO Emission (Using Natural Gas as Fuel)

The most recent data of this sort was published by Blakeslee and Selker[76] for new and existing tangential, coal-fired utility boilers. This data is summarized in Figures 5-16 and 5-17. It is felt that these costs should approximate those for units of other designs, firing different fuels. The bands of area explicitly show the ranges of the data, and that retrofitting existing units is always more expensive than if the modification was included in the original design.

Further data exists on retrofitting a 15 percent flue gas recirculation system on existing wall-fired units operating on gas and oil. Teixeira[98] states that a probable upper cost limit would be $4.2/KW for a 480 MW retrofit, occasioned by extensive hardware changes and increased operating costs. Barr and James[13], for the same modification but on a 750 MW gas-fired unit, reported a cost of $1/KW. It involved a far less complex conversion program, and is considered the probable lower cost limit.

Most investigators agree that low excess air operation decreases operating costs by increasing boiler efficiency slightly. This technique has become standard operating procedure on most existing utility and industrial boilers. Cost savings afforded by LEA operation are usually supplanted by additional, concurrent modifications.

As mentioned in Section 5.4.2.2, a commonly-used burner modification for biased firing is enlargement of the atomizing orifices to accomodate the higher fuel flow rate caused by out-of-service burners. Southern California Edison Company experienced a cost of approximately $5000 per unit for this modification. This technique is generally limited to new and existing gas- and oil-fired plants. Implementation on existing coal units would necessitate more elaborate and expensive changes in fuel handling equipment if they are not currently capable of firing dual fuels. Additional cost estimates were given by Berkau and Lachapelle[81].

Increased operating costs are normally associated with combustion modifications, but published data is scarce. One set of data, published by Blakeslee and Selker[76] is shown in Table 5-5. It should be noted that although the total annual cost increases with boiler size, the operating cost on a KWHR basis decreases.

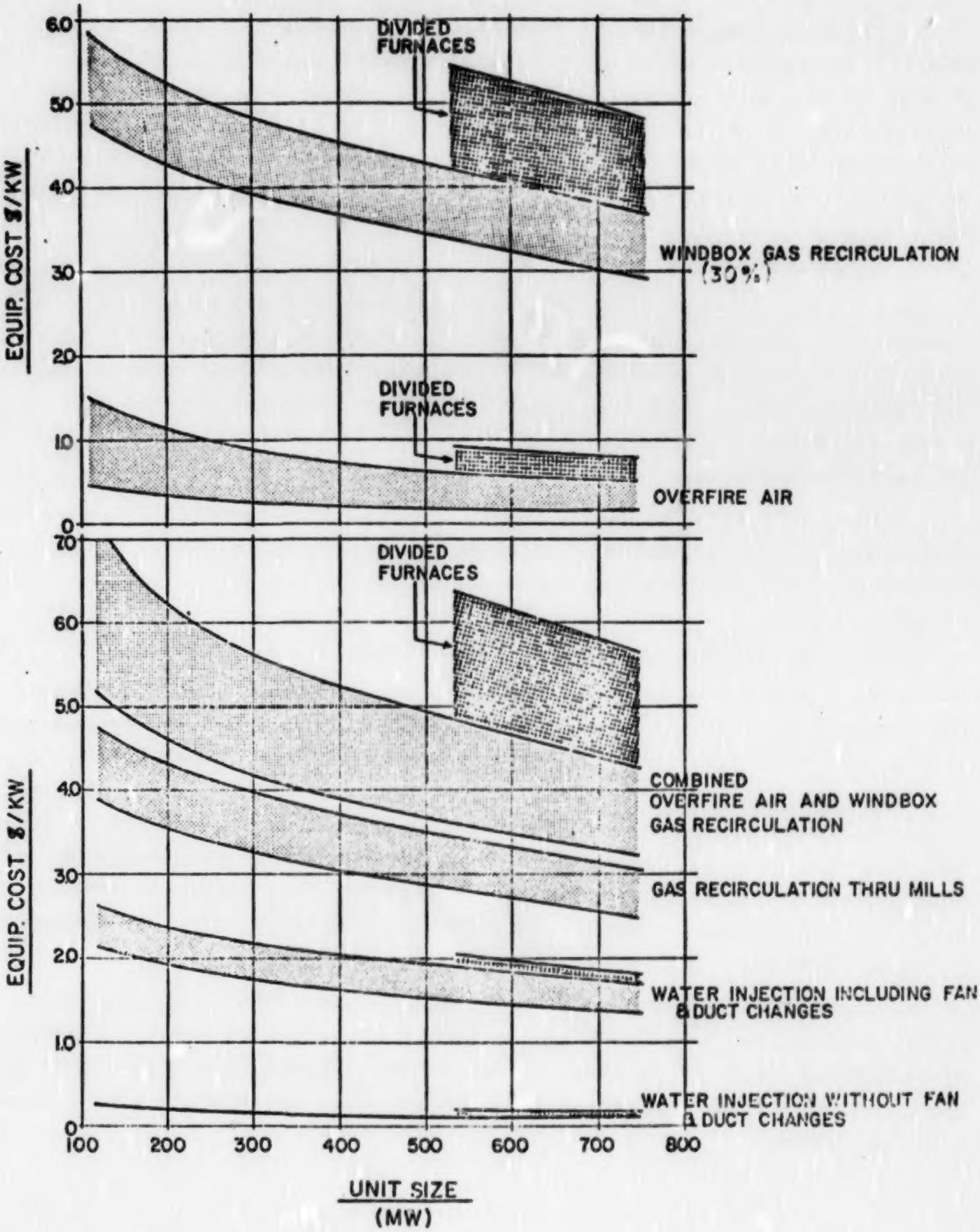


Figure 5-16. Equipment Costs of NO_x Control Methods for Existing Coal-Fired Units (Heat Transfer Surface Changes not Included)

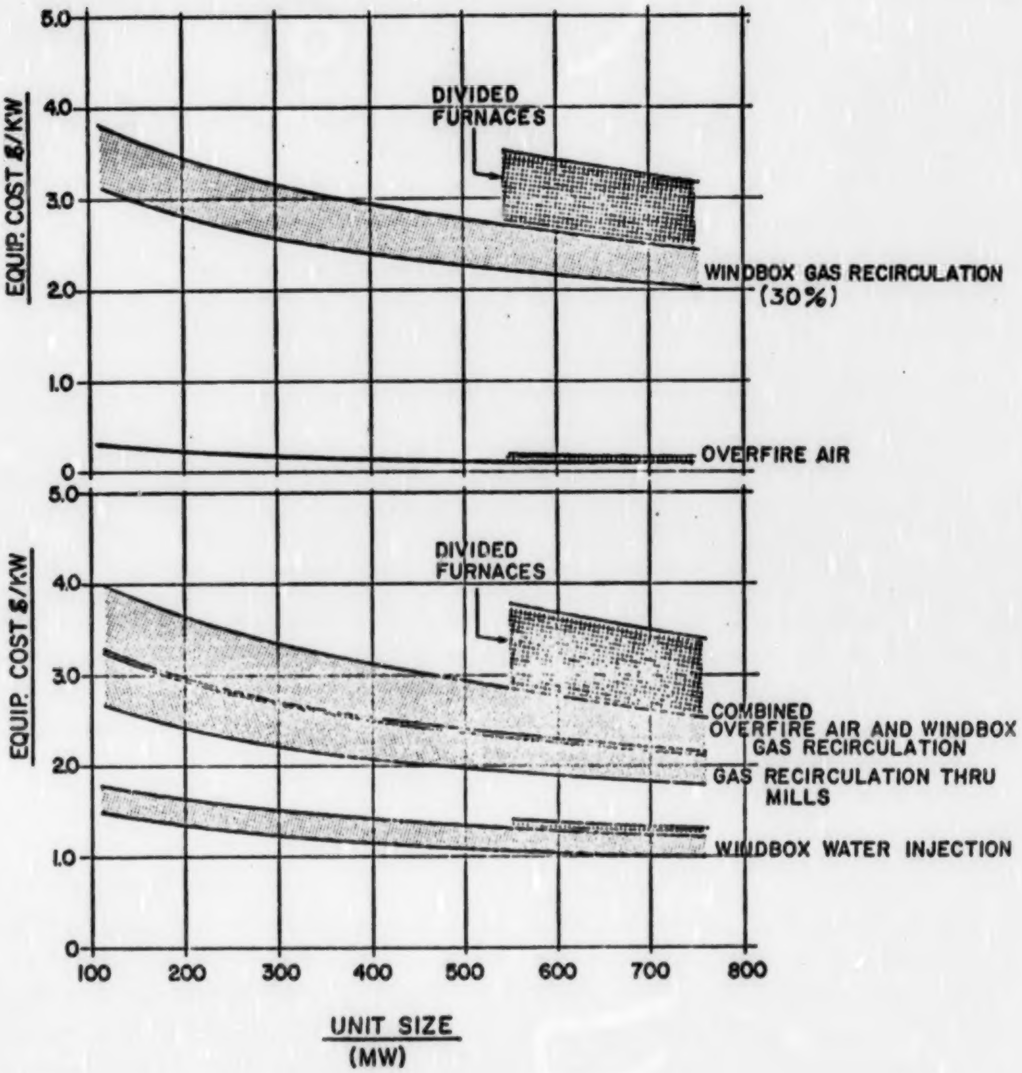


Figure 5-17. Equipment Costs of NO_x Control Methods for Existing Coal-Fired Units (Heat Transfer Surface Changes not Included)

TABLE 5-5

1973 OPERATING COSTS OF NO_X CONTROL METHODS FOR NEW COAL-FIRED UNITS (TANGENTIAL); SINGLE FURNACE

Control Method		Overfire Air (20%)			Windbox Flue Gas Recirc. (30%)			Combination of 1 and 2			Coal Mill Flue Gas Recirc. (17%)			Water Injection		
MW Rating		100	450	750	100	450	750	100	450	750	100	450	750	100	450	750
Equipment Costs	$10^3\$$	31	63	90	350	1185	1650	375	1248	1800	300	1015	1425	160	560	825
Annual Fixed Charge	$10^3\$$	5	10	14	56	190	264	60	200	288	48	162	228	26	90	132
Additional Annual Fuel Cost	$10^3\$$	---	---	---	---	---	---	---	---	---	---	---	---	147	660	1099
Additional Annual Fan Power Cost	$10^3\$$	---	---	---	21	95	158	21	95	158	22	100	166	13	58	97
Total Annual Cost	$10^3\$$	5	10	14	77	285	422	81	295	446	70	262	394	186	808	1328
Operating Cost	Mills/KWHR	0.009	0.004	0.003	0.143	0.117	0.104	0.150	0.121	0.110	0.130	0.108	0.097	0.344	0.332	0.327 (G

Based on:
A. Delivered and erected equipment costs (± 10% accuracy). Excluding contingency and interest during construction.
B. 5400 HR/YR at rated MW and net plant heat rate of 9400 BTU/KWHR.
C. 50¢/10^6BTU coal cost.
D. $250/HP fan power cost, or $40/HP per year.
E. Annual fixed charge rate of 16%.
F. Operating costs are ± 10%.
G. Does not include cost of water piping in plant or cost of makeup water.

Base unit operating costs* for coal fired power plants excluding SO_2 removal systems.

Unit Size	MW	100	450	750
Operating Cost	Mills/KWHR	16.2	13.5	12.6

*Includes 1973 capital costs, labor, maintenance, fuel costs +20% contingency +17% interest during construction.

Essentially no data were readily available on the costs of implementation of combustion modifications on industrial boilers. It is anticipated, however, that such information will be diffuse and generally difficult to interpret and correlate, directly reflecting the wide spectrum of equipment design and normal operating conditions.

5.5 SUMMARY

From a systems perspective, the most significant developments in combustion modification technology since the Esso study are the increasing dependence on coal with the concomitant problem of fuel NO_X, and the formation of the combustion control program to develop the technology for NO_X control. In this section we have reviewed the problem of NO_X formation and the status of the component areas of the program directed at attacking the problem in order to identify developmental requirements for systems planning.

Although preliminary identification of chemical and physical mechanisms in thermal and fuel NO_X formation has been made, there is still an inadequate understanding of the relationship between fuel and equipment parameters and NO_X formation. Accordingly, the program to devise and test modification techniques is broadly based with simultaneous focus on fundamental studies, pilot scale testing on laboratory scale equipment, and full scale testing on commercial equipment. The overall short term objective of the program is to develop verified, cost effective techniques for retrofit modification of existing units. Most of the significant reductions in NO_X emissions have resulted or directly benefited from the efforts of this program. The overall long term objective of the program is to develop optimized control concepts for new units and to define minimum attainable emission levels. For both the short and long term studies, the priority development area is combustion modification techniques for control of fuel nitrogen conversion to NO.

The fundamental studies area of the combustion control program is primarily a long term effort to develop a basic understanding of NO_X formation. Since the Esso study, the emphasis has been on modeling the individual key phenomena involved in NO_X formation. Significant advances have been made in the areas of fluid flow solution, thermal NO_X kinetics, gaseous hydrocarbon oxidation, and thermal radiation. These results have had the short term yield of aiding in evaluating data from pilot and full scale tests, and in suggesting ideas for combustion modification techniques. Only preliminary advances have been made in the modeling of fuel NO_X and of the effects of turbulent mixing on hydrocarbon and NO reaction rates. These are both considered priority R&D areas. In the long term, the individual models for the key phenomena will be coupled, refined, and verified by correlation with test data to ultimately yield predictions of NO_X formation.

Combustion modification testing is conducted within the fuels R&D, process R&D, and field testing component areas of the combustion control program. These efforts have resulted in operational guidelines for use by industry in NO_X control through combustion modifications. Stationary source combustion or other operating modification techniques, their major NO_X reducing effects, and problems usually associated with them are summarized in Tables 5-6a. and b.

For utility and industrial boilers, the effectiveness of these modifications on NO_X reduction depend on a large number of factors. These include boiler geometry and cleanliness, burner design, spacing, and state of repair, load, and, most importantly, the fuel itself. All of the techniques discussed are effective to varying degrees on gas firing due to suppression of thermal NO_X production. The contribution of fuel NO_X reduces both the number of cost-effective techniques and net NO_X reduction figures attained by them for oil and coal fuel. To date, published data have cast doubt on the NO_X-reduction effectiveness of temperature-reduction schemes, such as flue gas recirculation, for combustion systems firing coal and heavy fuel oils, both of which contain nitrogen-bearing compounds. By the same token, off-stoichiometric combustion has been shown to reduce both thermal and fuel NO_X, but more data is required, especially for coal combustion.

TABLE 5-6a

MODIFICATION TECHNIQUES FOR UTILITY AND INDUSTRIAL BOILERS

Modification Techniques	Major Effect	Major Potential Problems
Low Excess Air Operation	Oxygen concentration reduction (Fuel-rich burner operation)	Increased furnace slagging Nearer smoke threshold
Off-Stoichiometric Combustion	Fuel-rich burner operation Reduced residence time of fuel at peak temperature Peak temperature reduction	Flame instability, smoking Higher convective section temperatures
Flue Gas Recirculation	Peak temperature reduction	Flame instability Boiler efficiency reduction
Reduced Air Preheat Operation	Peak temperature reduction	Boiler efficiency reduction
Load Reduction	Peak temperature reduction	Boiler efficiency reduction
Water, Steam Injection	Peak temperature reduction	Boiler efficiency reduction Increased corrosion
Equipment Modifications:		
● "Low NO_x" Burner	Provides off-stoichiometric combustion at burner Readily fuel/air adjustable Fuel-flexible	Retrofit problems: Compatibility with existing furnace Conversion investment costs Boiler downtime
● Boiler Wash	Maintains rated heat transfer rate	Boiler downtime Maintenance expense
● Widen Burner Spacing	Reduce peak temperature Reduce interference between burner flames	New unit design only; retrofit cost prohibitive
● Tangential firing (as opposed to wall-firing)	Peak temperature reduction Slower, more controlled, combustion	New unit design only

TABLE 5-6b

MODIFICATION OR CONTROL TECHNIQUES FOR STATIONARY I.C. ENGINES AND GAS TURBINES

Modification or Control Techniques	Major Effect	Major Potential Problems
	Reciprocating I.C. Engines	
Speed vs. Stoichiometry	With speed, NO increases under fuel-rich and decreases under fuel-lean conditions	Retrofit difficulties; inability to meet load demand
Decreased Torque Load (at constant speed)	Peak temperature reduction	Retrofit difficulties; inability to meet load demand
Decreased Air Manifold Temperature	Peak temperature reduction	Efficiency reduction
Increased Valve Overlap	Peak temperature reduction	Fuel economy reduction Applicable only to 4-cycle engines
Exhaust Gas Recirculation	Peak temperature reduction	Intake manifold fouling Additional control system Operational difficulties Efficiency reduction
Catalytic Converter, Ammonia as Reducing Agent (Post Combustion Control Method)	Reduction of NO, NO_2 to N_2 and O_2	Expensive Current catalysts sensitive to impurities in fuel
Water Injection	Peak temperature reduction	Increased maintenance; additional equipment for water handling
Precombustion chamber	Peak temperature reduction; O_2 starvation	Costly for retrofit

TABLE 5-6b (Concluded)

Modification or Control Techniques	Major Effect	Major Potential Problems
Gas Turbines		
Lean-Out Primary Zone by Modifying Combustion Chamber Design	Peak temperature reduction Reduced residence time of fuel at peak temperature	Less control over flame stabilization Less control over lower lean extinction performance
Water injection	Peak temperature reduction	Reduced efficiency Increased maintenance Additional equipment for water handling
Exhaust Gas Recirculation	Peak temperature reduction	Reduced efficiency Additional control system Operational difficulties

SECTION 6

ASSESSMENT OF R&D REQUIREMENTS

Esso concluded the systems study of stationary source NO_x emissions with a detailed R&D plan in four categories:

- Combustion process studies
- Combustion flue gas treatment
- Noncombustion process studies
- Supporting studies

The priority R&D recommendations were in the combustion process studies area, which was further subdivided into Basic and Supporting Research, and Applied R&D. As described earlier, the EPA subsequently formulated an R&D program which incorporated and expanded upon the Esso R&D plan. The results of these and related efforts were reviewed in Chapters 4 and 5, and the purpose of this chapter is to suggest R&D topics for areas identified in that review as requiring further study.

Due to the limited scope of the present study, no attempt has been made to specify R&D schedules or level of funding. That approach would require a more comprehensive review of ongoing programs as related to future requirements. A qualitative ranking of priorities into high, medium, and low categories is given based on probable impact on emissions for short-term programs, or, low-risk development for long-term programs.

6.1 COMBUSTION MODIFICATIONS

The overall trend in combustion modification R&D is to establish the relationships between fuel or equipment parameters and the operating conditions promoting NOX formation. Within this context, the emerging emphasis is on NO_x formation in coal fired units, and on control technology for area sources such as industrial combustion equipment, IC engines, and commercial or domestic heaters.

Table 6-1 summarizes ongoing EPA research (for stationary sources) by relating R&D study areas to specific contracts in the four areas of fundamental studies, fuels R&D, process R&D, and field testing and survey. There are additional relevant programs which are funded outside the EPA. These are primarily in the areas of fundamental studies and field testing. Also, between the EPA stationary source and mobile source programs, there are mutually beneficial areas that could be exploited.

The following four sections give suggested additions or extensions to the ongoing programs listed on Table 6-1. These R&D recommendations are further summarized in Table 6-2.

6.1.1 Fundamental Studies

Several of the more pressing problems in the fundamental studies area are the development of a basic understanding of fuel NO_x conversion, the need to relate fundamental studies to measured performance, and, more broadly, the development of scaling laws for generalizing pilot-scale test results. R&D suggestions relating to these areas, as well as the ongoing long-term programs, are given below in the context of the fundamental studies framework presented in Section 5.

Fluid Flow Solutions: There exists an extensive library of computer codes for flow prediction in a variety of geometries. The priority items in this area are, therefore, not with basic code development, but with testing of auxiliary relations for use in existing codes.

Medium priority:

- Continue use of 2-D recirculating code to test models for swirl, radiation, kinetics, etc., and to increase numerical stability and reduce running time.
- Apply 2-D jet flame boundary layer codes to test turbulence models, mixing effects on kinetics, 2ϕ flow models, luminous radiation effects, etc. Supplement with experimentation on these effects.

Low priority:

- Initiate use of pressure-velocity method for 2-D recirculating flow to gradually replace the stream function-vorticity method.
- Assess use of pressure-velocity method for 3-D flow for obtaining flow patterns and scaling relations.

Thermal NO_x: The high priority items in this area are covered by existing programs. Supplementary interest areas of medium priority are:

- Determine HCN emissions from fuel-rich flames.
- Utilize results of Esso/Ultrasystems study in a realistic flow situation, such as 2-D jet flames, with full diffusional effects.

Fuel NO_x and 2ϕ Droplet/Particulate Combustion: Rocketdyne is currently doing a high priority exploratory study of the most important aspects of fuel

TABLE 6-1

SUMMARY OF COMBUSTION RESEARCH SECTION PROGRAM

FUTURE R & D AREAS / EPA PROGRAMS	Chemical Reactions of Fuel Constituents NAR Rocketdyne	Reaction Chemistry of Pollutant Formation ESSO & R & E	Math. Model of Pollutant Formation Process Ultrasystems	Aerodynamic Phenomena in Pollution Control UARL	Flame & Thermal Interactions NASA-JPL	Computerized Estimation of Rate Constants SRI	Pollutant Emission Control Through Catalysis BMI	Change in Flame Species Conc. on Poll. U Arizona	Characterization of Emission Types & Levels EPA-CRS	Chemical Reactions of Soot & Related Poll. MIT	Fundamental Research Support Aerospace	Design Principles for Liquid & Solid Fuels IFRF	Combustion Modification Control for Coal B & W	Evaluation of Comb. Control Tech. EPA	Design Principles for Liquid Fuels NAR-Rocketdyne	Design Principles for Gaseous Fuels IGT	Comb. Mod. for Ind. Package Boilers Ultrasystems	Development & Application of Comb. Control EPA	Evaluation of Low Pollution Catalytic Bur. EPA	Non Polluting Catalytic Combustors	Staged Combustion for Tang. Coal Fired Boil. C.E.	Prelim. Eval of Comb Mod for Wall Coal Fired B. USBM	Staged Comb for Wall Coal Fired Boilers TVA	Design, Trends & Problems W/Comb Cntrl of Ind B. BMI	Application & Dev. of Automotive Tech Aerospace	Anal of Comb Cntrl for Gas & Oil Fired Util B. Aerospace	Field Testing Power Gen. Equipment ESSO R & E	Field Testing of Industrial Boilers KVB- ENGR	Char. of Prototype & Commer Comb Sys. EPA	Survey of Ind. Furnaces	Identification & Char. of Waste Fueld M.W. Kellog	Development of Probe Techniques Arnold Res.	In-Situ Measurement Methods Dev. NAR-Rocketdyne
FUNDAMENTAL STUDIES																																	
• Fluid Flow Solutions			●	●				●		●																							
• Thermal NO$_x$	●	●	●	●		●	●	●		●																							
• Fuel NO$_x$	●							●		●																							
• Turb. Visc.				●						●																							
• Turb Mixing/Kinetic Coupling								●		●																							
• Radiation				●																													
• 2φ Droplet, Part. Comb.	●							●		●																							
• Hydrocarbon Combustion	●	●	●	●		●		●		●																							
FUEL R & D																																	
• Staged Comb.													●																				
• Burner Opt. for Fuel N$_2$												●			●																		
• Radiation Measurement												●																					
• Corrosion & Fouling																																	
• Coal Types																																	
• Synthetic Fuels																●																	
• Oil Atomization															●																		
• Optimized Modification													●	●		●																	
• Optimized Burner Design																●																	
PROCESS R & D																																	
• Corrosion Studies																					●		●										
• Catalytic Comb.																			●	●													
• Staging, Overfire Air																					●	●	●										
• Intermed. CBN Compounds																																	
• Optimized Modification & Area Sources																	●	●				●		●	●	●							
FIELD TESTING																																	
• Corrosion for Var. Coals																											●						
• Modification for Pulv. Coal																											●	●					
• Emission from I.C. Engines																													●				
• Capability of Gas & Oil Fired Equipment																											●	●	●	●			
• Modifications for Other Coal Comb.																												●					
• Other Fuels																															●		
• Instrumentation																																●	●

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TABLE 6-2

RECOMMENDED R&D

R&D Area	Subheading	Program	Priority	New Program/ Continuation	Reasoning
Fundamental Studies	Fluid Flow Solutions	Continue 2-D code development	Medium	Continuation	Incorporate current auxiliary models
		Apply 2-D jet flame boundary layer codes	Medium	New	Test turbulence model, mixing effects on kinetics, 2ϕ flow models, luminous radiation effects
		Pressure-velocity codes for 2-D recirculating flow	Low	New	Utilize current technology
		Pressure velocity method for 3-D	Low	New	Obtain flow patterns and scaling laws
	Thermal NO_x	HCN emission in fuel-rich flames	Medium	Continuation	Determine potential as a pollutant
		Esso-Ultrasystem codes with diffusional effects	Medium	Continuation	Extend findings to practical flow situations
	Fuel NO_x	Volatilization + pyrolysis models	High	Continuation	Extend Rocketdyne findings into independent studies
		Post-pyrolysis heterogeneous carbon burnout models	High	Continuation	" " "
		Couple particulate combustion with luminous radiation	High	Continuation	" " "
		Synergism of NO_x and SO_2	High	Continuation	" " "
		HCN	High	Continuation	Determine potential as a pollutant
	Turbulent Viscosity	2 Equation turbulence models in recirculating 2-D flow code	Medium	New	Utilize current technology
		Nonisotropic turbulence models for swirling 2-D boundary layer jet flames	Medium	New	Verify models in simple flow geometries
	Mixing Effect On Reaction Rates	Experimental and analytical study of turbulent premixed and diffusion flames - simple geometry	High	New	Develop models for effect of mixing on reaction rates
	Thermal Radiation	Coordinate existing radiation models with fuel nitrogen study	Medium	Continuation	Utilize findings of Rocketdyne study
Fuels R&D	Off-Stoichiometric Combustion	Determine optimum design parameters for staged firing with coal	High	New	Present research indicates this to be most viable NOx control technique for coal combustion
	Burner Optimization for Fuel NO_x	Determine optimum design parameters for low NO_x burner	High	New	Effective option for fuel nitrogen control

Table 6-2. Continued

R&D Area	Subheading	Program	Priority	New Program/ Continuation	Reasoning
Fuels R&D continued	Burner Optimization for Thermal NO_x	Determine optimum design parameters for low NO_x burner using low nitrogen fuels	High	New	May be effective option for thermal NO_x as well
	Radiation Measurement	Determine realistic radiation heat fluxes for coal, oil and gas	Low	New	Input to radiation models
	Corrosion & Fouling	Determine ability to do subscale corrosion and fouling studies of NO_x control schemes	High	New	Very important factor to determine applicability of controls
	Coal Type	Systematically test different types of coal - effect of properties on NO_x emissions	Medium	New	Determine applicability of control techniques over spectrum of coal types
	Synthetic Fuels	NO_x emissions from fuels and equipment conversion problems	Medium	Continuation	Necessary to identify problems of future fuels
	Oil Atomization	Determine effects of fuel atomization technique on NO_x	Medium	Continuation	Potential control technique; coordinate with optimum burner design
	Optimization Modification	Determine optimized modification schemes for each fuel and process	Medium	Continuation	Necessary for immediate retrofit
Process R&D	Corrosion Studies	Perform full scale studies of NO_x control techniques on corrosion and fouling	High	Continuation	Full scale testing will yield the ultimate data
	Off-Stoichiometric Combustion	Expand full scale testing for staging on coal combustion	High	Continuation	" " "
	Intermediate Bound Compounds	Determine fate of chemically bound nitrogen in full scale equipment	Low	New	Determine if there really is a problem in full scale equipment
	Optimized Modification and Areas Sources	Continue to expand ongoing programs to determine optimized schemes; especially area sources	High	Continuation	Determine applicability on full scale equipment
	Catalytic Combination	Exploratory work to determine full potential	Low	Continuation	Potential long range option
Field Testing	Pulverized Coal-Fired Equipment	Determine current stationary source control technology (CSST)	High	Continuation	Coal-fired equipment has highest priority
	Corrosion & Slagging	Determine effects of CSST on corrosion and fouling	High	Continuation	Additional data very important
	Area Sources	Determine CSST for area sources	High	Continuation	Field data lacking

Table 6-2. Concluded

R&D Area	Subheading	Program	Priority	New Program/ Continuation	Reasoning
Field Testing continued	Alternate Fuels	Exploratory work to determine nature and extent of problem	Low	New	Extent of use and NO_x problems are unknown
Flue Gas Treatment	--	Review state of the art	Low	New	May be required for secondary cleanup
Alternate Processes	Synthetic Fuels	Determine NO_x problems	Low	New	Fuels may be extensively used in 1980-2000
	Advanced Power Generation Cycles	Determine NO_x problems	Low	New	" " "

nitrogen conversion and droplet/particulate combustion. As preliminary results become available, these could be used as the basis for further, independent, high priority studies:

- Volatilization and pyrolysis models of single droplet or particulate flow. Extend to one or 2-D combustion of a cloud of particles.
- Post-pyrolysis heterogeneous carbon burnout models.
- Coupling of particulate combustion results with luminous radiation modeling.
- Incorporate Rocketdyne findings in University of Arizona study on synergism of NO_x and SO_2.
- Establish level of HCN emission due to fuel nitrogen intermediaries.

Turbulent Viscosity: A medium priority effort in turbulence modeling would benefit the use of computer codes to predict combustion flow fields.

- Test the two-equation turbulence models in a recirculating 2-D flow code.
- Test nonisotropic turbulence models for swirling 2-D boundary layer jet flames.

Effects of Mixing on Reaction Rates: This is a high priority area as flame zone mixing can dominate the reaction rates for combustion and NO_x formation. For developing fundamental understanding, a closely coordinated experimental and analytical study of turbulent premixed and diffusion flames in simple geometries would be most beneficial.

Thermal Radiation: A continuing effort should be made to incorporate into existing radiation models results obtained from the fuels nitrogen study and efforts in other auxiliary areas.

6.1.2 Fuels R&D

This research area derives its nomenclature from the historical investigations on pilot scale versatile combustion equipment for each "fuel" type, coal, oil and gas. Basically, it is concerned with subscale experimental studies on equipment with the flexibility to vary burner parameters or to study in staging, overfire air, flue gas recirculation, air preheat, etc. Based on the results of the present study, the following areas of pilot scale activity are identified as needing further definition.

	Priority
• Off-stoichiometric combustion	High
• Burner optimization for fuel nitrogen	High
• Burner optimization of thermal nitrogen	High
• Radiation measurements	Low
• Corrosion, fouling tests	High
• Systematic testing of coal types for effects of chemically bound nitrogen and S/N ratio	Medium
• Synthetic fuels	Medium
• Oil atomization studies	Medium
• Optimum modification scheme for each fuel and process	Medium

An indication of priorities, high, medium or low is given for each of the study areas. High priority areas include those associated with NO_x emissions from coal combustion, fuel nitrogen problems, problems associated with area sources, or where vital information on any important subject is lacking. Medium priority attention is given to those areas where some information is available but additional data would be very helpful, or areas that need work but are not quite as essential for controlling the major NO_x sources. Low priority items generally include long range problems and those areas that would contribute additional understanding if funds were available.

Each of these fuel R&D areas will be briefly described in the following paragraphs. Further work is necessary to define exactly what should be done in each of these areas. As was noted on Table 6-1, the current R&D program covers most of these areas to some extent but is limited by available funds.

Off-Stoichiometric Combustion (High Priority)

The emphasis in this area should be on the application of off-stoichiometric firing techniques to coal combustion. Tests are needed to determine the effects of the following parameters on staged combustion:

- First stage air/fuel ratio
- Residence time to point of injection of secondary air
- Direction and method of injection of secondary air
- Heat absorption between stages
- Burner variables (swirl, velocities, etc.)

Studies with oil and gas firing would also be of interest.

Burner Optimization for Fuel Nitrogen (High Priority)

Burner design principles need to be identified to achieve low NO_x emissions for fuels with chemically bound nitrogen. The optimum burner design would achieve the same effect as staging through local combustion aerodynamics. It is realized that a universal burner for all applications (boilers, furnaces, etc.) is not practical, but development would benefit from identification of general design principles. A burner design for utility boilers may be the best focal point for this project.

Burner Optimization for Thermal Nitrogen (High Priority)

A different approach to burner design may be desirable when fuel nitrogen conversion is not of concern. Although the use of natural gas as a fuel for utility boilers seems to be on the decline, its combustion will remain a significant factor for emissions from area sources. Therefore, burner design principles should be developed for low NO_x distillate oil and gas burners. Devising an optimized burner design for typical home-heater size combustion systems would be on approach.

Radiation Measurements (Low Priority)

In order to provide a realistic heat transfer model of the combustion process, data on radiation heat fluxes from gas, oil and coal flames is essential. Tests could be performed in a versatile subscale piece of equipment. However, care must be taken to ensure that wall heat fluxes resemble those in full scale equipment.

Corrosion and Fouling Test (High Priority)

Since corrosion and slagging problems are of vital concern to utility boiler operators, it is essential that the detrimental effects of the various NO_x control schemes be determined. The possibility of doing accelerated subscale corrosion and fouling tests should be investigated as it would, no doubt, be much cheaper than full scale testing. Many investigators, however, contend that subscale corrosion tests are not reliably scaled to full scale equipment. Exploratory tests would be helpful in this regard.

Systematic Testing of Coal Types (Medium Priority)

There are many varieties of coal, due to differing proportions of nitrogen, sulfur, carbon-hydrogen ratio, ash, heat capacity and other properties. It is very likely that a wide range of NO_x emission magnitudes can result from use of these coals. Thus, a systematic study in subscale apparatus is recommended to determine the relative effects of percentage fuel nitrogen, sulfur nitrogen ratio and other coal properties on NO_x emission.

Synthetic Fuels (Medium Priority)

As was noted in Section 3, considerable research activity will take place in the next decade aimed at developing synthetic gaseous and liquid fuels from coal, and oil from oil shale and tar sand. Studies should be initiated to evaluate the possible NO_x emissions from the combustion of such exotic fuels, as well as the concomitant problems (i.e., load derating) of converting existing plants to these fuels.

Oil Atomization Studies (Medium Priority)

An integral part of an oil-fired combustor design is the manner in which the oil is atomized. The atomization technique and the mixing patterns will greatly influence the formation of NO_x. It is recommended that the current studies at Rocketdyne be continued and possibly expanded.

Optimized Modification Scheme for Each Fuel Type and Application (Medium Priority)

As was noted in Table 6-1 there are several ongoing studies to determine the optimum NO_x control scheme for each fuel and application. It is felt that these types of studies, including periodic evaluation of the status of the schemes, are valuable and should be contined. Perhaps greater emphasis should be placed on determining optimized NO_x reduction schemes for area sources, such as industrial boilers, furnaces and domestic and commercial heating systems, using versatile, although not necessarily subscale, equipment.

6.1.3 Process R&D

Process R&D is concerned with the full scale implementation of one or more control schemes. Recommended R&D study areas and priorities are as follows:

	Priority
● Corrosion studies	High
● Off-stoichiometric combustion (staging)	High
● Intermediate chemically bound nitrogen compounds	Low
● Optimized modifications and area sources	High
● Catalytic combustion	Low

Each of these areas will be briefly discussed.

Corrosion and Fouling Studies (High Priority)

Two full scale corrosion and fouling studies will soon be implemented (conducted by Combustion Engineering and the TVA). It is recommended this area of research be greatly expanded. A short study should be undertaken to determine the most effective means of averting the corrosion and slagging problems associated with NO_x control schemes. It would be highly desirable to be capable of performing subscale corrosion tests as was suggested in Section 6.1.2. An accelerated corrosion and slagging program in full-scale units should in any case, be promoted.

Off-Stoichiometric Combustion (Staging) (High Priority)

The findings from the fuels R&D staging studies should be applied to full-scale equipment to verify the results. The Combustion Engineering and TVA demonstrations will contribute valuable data, but will not provide much information regarding the optimum injection point. Although it is realized that full-scale testing is rather costly, it is recommended that additional studies be made.

Intermediate Chemically Bound Nitrogen Compounds (Low Priority)

As was discussed in Section 5, there remains some question as to the fate of chemically bound nitrogen. That is, whether it is converted to NO, N_2 or even HCN. It is proposed that the fate of these compounds in full-scale equipment be studied to determine how the conversion, especially conversion to HCN occurs. Results obtained should be coordinated with data from pilot-scale and bench-scale tests.

Optimized Modification and Area Sources (High Priority)

There are a number of ongoing programs in the process R&D area to determine or evaluate the optimum NO_x control scheme. These programs are of great value and should be continued. Although programs concerned with controls for coal combustion in utility boilers should not be de-emphasized, more effort should be placed on controls for area sources, such as industrial boilers and furnaces, commercial and home heating systems as well as IC engines and gas turbines. These studies may require more funds than are now available. A number of programs are in the planning stage at EPA[15].

Catalytic Combustion (Low Priority)

Catalytic combustion has been shown to be a potential low NO_x emission combustion mechanism. Some additional exploratory work to determine the full potential of catalytic combustion should be performed.

6.1.4 Field Testing

Field tests have been used in the past to determine current emission levels as well as the capability of existing equipment to produce lesser quantities of NO_x. Much of this work is currently underway or has been completed. However, the following areas require continuing or additional work.

	Priority
● Field testing of pulverized coal-fired equipment	High
● Corrosion and slagging for various types of coals	High
● Field testing of area sources	High
● Alternate fuels	Low

Field Testing of Pulverized Coal-Fired Equipment (High Priority)

This test work is an ongoing program at EPA, through Esso Research and Engineering, to determine levels of NO_x emissions and control capability of existing equipment. This work is providing significant background material and, in light of many utilities switching to coal in the near future, it is recommended that studies of this type be accelerated and expanded.

Corrosion and Slagging (High Priority)

The above field tests offer an excellent opportunity to determine the effect of selected control methods (i.e., low excess air, flue gas recirculation, and off-stoichiometric combustion) on slagging and corrosion for many types of boilers. This is being done through the Esso field test work on utility boilers and should be continued and expanded in light of fuel conversion activity.

Field Testing of Area Sources (High Priority)

There has been considerable emission survey work on industrial boilers and commercial and home heating systems. Additional statistically representative data for these sources would be worthwhile. Applying NO_x reduction techniques to them is the next logical step. In addition to this ongoing work, emissions and controls for other area sources such as industrial furnaces, stationary reciprocating IC engines, and gas turbines need to be defined. In particular, as was indicated in Section 3, there is a considerable range of NO_x emission estimates for IC engines. The importance of this source of emissions requires clarification.

Alternate Fuels (Low Priority)

Combustion of alternate fuels, particularly waste materials, may have an associated NO_x emission problem. Fuels such as pitch and waste products from chemical plants are used in process heaters or are directly incinerated. Some exploratory work should be performed to determine the nature and extent of the problem.

6.2 FLUE GAS TREATMENT

Since this area was not extensively explored in the present study, it is rather difficult to make valid R&D recommendations. Perhaps as a first step a study should be undertaken to thoroughly review the state of the art and recommend areas of future research. Some NO_x researchers interviewed during the course of this survey recommended additional flue gas treatment studies because this option may be needed as a secondary cleanup scheme to meet more stringent laws (especially as the population of sources increases). Flue gas treatment may be the most cost-effective technique for some of the smaller sources comprising area sources. At this time, however, combustion modification still seems to be the most promising alternative.

6.3 ALTERNATE PROCESSES

Some funds should be allocated for monitoring the status of and the developments in the longer range alternate power generation schemes. These may include the following:

- Synthetic fuels:
  - Methanol
  - Hydrogen
  - Garbage and waste
  - Low Btu and High Btu gas
  - Sewer gas
- Advanced power generation cycles
  - Fluidized bed boiler
  - Combined cycle

If, at some future time, one or more of these processes show promise, additional programs should be initiated to characterize any NO_x emission problems associated with such systems.

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